

RADIOACTIVE MATERIALS IN FOOD AND AGRICULTURE

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Radioactive mate..

CULTURE ORGANIZATION OF THE UNITED NATIONS

RADIOACTIVE MATERIALS IN FOOD AND AGRICULTURE



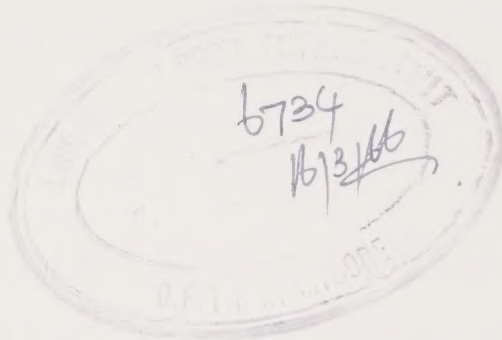
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RADIOACTIVE MATERIALS IN FOOD AND AGRICULTURE

Report of an FAO Expert Committee
Rome, 30 November-11 December 1959

FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS

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TABLE OF CONTENTS

Participants in the meeting	VII
Members of the Committee	VII
Consultants to FAO	VIII
Observers	VIII
FAO Secretariat	IX
Introduction <i>radioactive materials ✓</i>	1
I. Sources of <u>radioactive contamination</u>	4
II. General aspects of <u>food chains</u>	5
Soil and plant contamination	5
Contamination of animals and animal products	6
Contamination of fisheries products	6
Food consumption <u>habits</u>	7
III. <u>Strontium</u> <i>food contamination</i>	8
Behavior in soils and plants	8
Animals and animal products	8
Aquatic food chains	9
Diet composition	10
Illustrative survey levels in <u>diets</u>	11
IV. <u>Cesium</u>	11
Behavior in soils and plants	11
Animals and animal products	12
Aquatic food chains	12
Diet composition	12
Illustrative survey levels in diets	12

V.	<u>Iodine</u>	13
VI.	<u>Other radioactive substances</u>	13
VII.	Remedial measures	15
VIII.	Surveys of environmental <u>radionuclides</u> in food and agriculture	17
IX.	Recommendations	20

Appendixes

1.	Radioactive nuclides of importance in relation to potential contamination of human food supply	24
2.	Radioactive contamination of soils and crops	35
3.	Movement and behavior of radioactive materials in animals and animal products	66
4.	Radioactive contamination in fisheries products	79
5.	The effect of food consumption habits and processing on the content in human diets of some radioactive and stable isotopes, with special reference to strontium, calcium, cesium and iodine.	95

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Dr. R. A. SILOW	Chief, Atomic Energy Branch (<i>Convenor</i> for the Director-General).
Dr. K. K. P. N. RAO	Chief, Food Consumption and Planning Branch, Nutrition Division.
Mr. F. HOYOS	Technical Officer, Atomic Energy Branch.

The Committee had the advantage of attendance of other staff members of FAO at appropriate times. These representatives included: Dr. F. W. Parker, Mr. T. E. Ritchie, Dr. V. Ignatieff, Mr. W. Reese, Dr. R. O. Whyte, Dr. J. B. Harrington, Dr. K. V. L. Kesteven, Dr. N. R. Reid, Dr. H. Konigshofer, Dr. E. Lancelot, Mr. R. R. Joshi, Dr. D. B. Finn, Dr. G. L. Kesteven, Dr. J. G. Knoll, Mr. H. Rosa, Mr. W. A. Dill, Mr. T. Laevastu, Mr. M.A. Huberman, Mr. N. E. Holmes and Mr. W. Schulte.

INTRODUCTION

The FAO *ad hoc* Expert Committee on Radioactive Materials in Food and Agriculture met at the Headquarters of the Food and Agriculture Organization of the United Nations in Rome, 30 November – 11 December 1959. The Committee was established by the Director-General to advise on the nature and scope of work that should be undertaken by FAO in response to a recommendation of the Tenth Session of the European Commission on Agriculture in May 1958, and in response to a resolution of the Sixth Session of the United Nations Scientific Committee on the Effects of Atomic Radiation indicating that it would welcome detailed and continuing consideration on an international basis by FAO of the problems of radioactive contamination of food and agriculture, in order that the Organization may assist the United Nations Radiation Committee to carry out the responsibility assigned to it by the General Assembly for evaluating the effects of ionizing radiations on man and his environment.

With these objectives the FAO Expert Committee on Radioactive Materials in Food and Agriculture was assigned the following terms of reference:

- (a) to review present knowledge of the behavior of radioactive substances in terrestrial and aquatic food chains, including their levels in foods and agricultural materials, and practices which affect the radioactive contamination of human diets;
- (b) to indicate possible improvements in surveys of radioactive substances in foods and agricultural materials;
- (c) to make recommendations regarding research on food chains needed to further the understanding and reduction of the radioactive contamination of the diet of man.

Sir William Slater and Dr. C. L. Comar were unanimously elected Chairman and Vice-Chairman respectively. Dr. R. A. Silow served as Technical Secretary.

The Meeting was opened on behalf of the Director-General of FAO, Dr. B. R. Sen, and on behalf of the Deputy Director-General, Dr. N.C. Wright, who had participated in preparations for the meeting, by Dr. F. W. Parker, Assistant Director-General in charge of the Technical Department. In welcoming participants, he referred to the fact that from the beginning the development of the principles and applications of atomic energy was an international effort and that the impact of the various uses of atomic energy acknowledges no national boundaries. During this atomic age, general and local rises in environmental radioactivity are highly probable and man must learn how best to live with them and to minimize their effects. As the primary route of environmental radionuclides into man is the food chain, the importance of agriculture, fisheries and food in determining the radioactivity burden of man is obvious. The Committee had been established to enable FAO to provide its member nations with a consensus of expert opinion on the present status of knowledge of radionuclides in food chains and on survey and research programs needed to extend that information.

Professor Rolf Sievert, in his capacity as Chairman of the United Nations Scientific Committee on the Effects of Atomic Radiation, expressed on behalf of that Committee its appreciation of the co-operation extended to it by FAO in the Organization's sphere of responsibility.

The Committee considered the problems of radioactive contamination of the food chain in terms of food, agricultural and fisheries experience and knowledge. Emphasis was placed on matters of soil and plant contamination, although consideration and representation were given to animal products, marine biology, human dietary aspects and over-all survey and sampling problems. It was noted that, while most of the data came from relatively few countries, it was always kept in mind in development of the basic concepts, that due consideration must be given to all populations. It may also be noted that studies have been mainly concerned with contamination of recent years. While emphasis has naturally been placed upon assessment of present or near future conditions, there has

always been present the idea of assessment of conditions for any future situations.

This report contains a brief summary of the conclusions and recommendations of the Committee. More detailed information and conclusions will be found in the five technical appendixes. Working papers considered by the Committee will be published in a separate supplement.

I. SOURCES OF RADIOACTIVE CONTAMINATION

The various applications of atomic energy involve the formation of quantities of radioactive fission products and some activation products which present a potential problem in regard to the contamination of human food. Appendix 1 reviews the manner in which these arise and indicates broadly the relative importance of the various nuclides.

The predominant environmental contamination at present results from the escape into the environment of fission products from nuclear explosions. The isotopes of concern are those with high fission yield and moderate to long half-life for which the biological properties are such that they are readily taken up into the human food chain and into human beings. On this basis the predominant isotopes are I-131, Cs-137, Sr-89 and Sr-90.

One of the ways in which environmental contamination can arise is from an accident with a large nuclear reactor, and experience in the accident at Windscale, United Kingdom, in 1957, where 20,000 curies of I-131 together with some other isotopes escaped, shows that the volatile fission products I-131, Cs-137 and to a lesser extent Te-132 and Ru-106 were biologically the most important nuclides. In areas where the herbage contamination from I-131 was $1 \mu\text{c}/\text{m}^2$ the level in milk was about $0.1 \mu\text{c}/\text{liter}$. Somewhat higher relative milk contamination could arise under summer conditions but in winter the level would be likely to be very small. In contrast to iodine, the contamination of foodstuffs by strontium isotopes was nowhere serious.

It appears from considerations of fission yield, half-life, and the physical and biological properties of the various nuclides, that among the fission products I-131, Cs-137, Sr-89, Sr-90 and to a lesser extent Te-132 and isotopes of ruthenium, are biologically the more important.

In addition to the fission products, many radioactive materials are produced by neutron activation. In nuclear explosions, for instance, the important radioactive isotope C-14 is produced from the reaction of neutrons with nitrogen. In nuclear reactors, various activation products arise, notably Co-60, Fe-55, Fe-59, Cr-51, Cu-64, Ta-182 and Zn-65, which may be released to the environment as corrosion products in water, and in some instances certain of them are produced in nuclear explosions. Fission products and activation product nuclides other than those listed in this general review may also become of importance in certain circumstances.

The use of radioactive isotopes in scientific research, medicine, industry and agriculture also gives rise to potential contamination from the discharge of waste materials into sewage systems and rivers.

The fuel used in nuclear reactors must periodically be chemically processed in order to remove the fission products which are then held as stored wastes. In this process small quantities of the volatile fission products and the noble gases escape to the atmosphere. It is also usually necessary to discharge from such plants dilute liquid radioactive wastes which are so voluminous that they cannot be stored. In these circumstances, the fission products and activation products mentioned above may be of importance as well as some additional isotopes such as Ce-144.

Although large quantities of radioactivity are involved in the various applications of atomic energy, it must also be recognized that they must be seen against a background of the widespread distribution in the surface soils of the numerous naturally-occurring radioactive elements in the uranium, thorium and actinium series and potassium and other radioactive elements. These also enter the human food chain and are present in the human body.

II. GENERAL ASPECTS OF FOOD CHAINS

Soil and plant contamination

Plant material may become contaminated from the atmosphere by fission products that do or do not become diluted with soil substances. Direct contamination may occur on leaves, fruits and seeds (foliar and floral absorption). Depending on the nature of the plant,

material not retained in this way may be partly absorbed by the basal parts and surface roots of the plant (plant-base absorption). Material also may enter the soil and then the plant via the roots. The soil route of entry, in addition to leading to dilution, contrasts with direct mechanisms of absorption, in that material deposited before the plant develops can enter the plant. Soil entry is unimportant with short-lived nuclides. If the soil is undisturbed, fission products will move downward only slowly. Surface rooted crops are therefore likely to absorb the largest amounts from soil deposits. The extent to which different fission products are absorbed by plants depends on many factors. Radionuclides contaminating plants may reach man directly by way of foods of plant origin or indirectly through animal products.

Contamination of animals and animal products

Animal products are an important link in the food chain by means of which radionuclides reach the human population because the grazing animal is an effective collector of contamination via plants. The quantitative relations that exist between the amounts of radionuclides that enter the animal and the amounts that become deposited in the tissues and secretions, particularly those used for food, are therefore of particular interest. There are many factors that influence the degree of exposure of farm animals to radionuclides, e.g., metabolic properties of the specific nuclide and feeding and management practices. These factors must also be taken into account when interpreting the results of surveys. The effect of exposure to radionuclides on the well-being of animals is not considered in this report because it will be of minor practical importance as compared to that of man even though grazing animals will generally acquire higher body burdens than will human populations.

Contamination of fisheries products

Fisheries products contribute only a small fraction of radioactive contamination to the human diet. This is true even for the Pacific Ocean which, because of close-in fallout, has widespread radionuclide concentrations up to about ten-fold those of other oceans. It is em-

phasized that organisms in the marine and freshwater environment, especially the primary producers, can concentrate certain elements to a high degree; thus specific neutron activated nuclides may be much more important in aquatic than terrestrial food chains. A real difference in contamination levels of the marine environment tends to be eliminated by processes of horizontal dispersion and vertical mixing.

Food consumption habits

In view of the fact that various dietary components are contaminated to different degrees, it is necessary to assess the contribution of the human diet as a whole to the human body burden. It is, therefore, necessary to have reliable estimates of total food and fluid consumption of the population under consideration, and analyses of representative samples of at least the most important dietary sources of a particular nuclide.

It is difficult to collect reliable estimates of food consumption, but much useful information on the subject is available in certain countries and in FAO. It is important to utilize this knowledge and experience in making plans to monitor food supplies or to make estimates of the dietary intake of any nuclide.

There are two main sources of data on food consumption patterns and levels, the food balance sheet and the dietary survey. The first is an estimate of the *per caput* supplies of foods available at the retail level for human consumption in a country as a whole and is mainly derived from records of production and trading. The second is a direct investigation of the food consumption of samples of different groups of the population. Such data are usually collected on a family basis.

The food balance sheet gives a broad indication of consumption patterns in different countries, but cannot usually be used satisfactorily as a basis for detailed investigations. The family dietary survey can provide more detailed information, but such information can be misleading if care has not been taken to survey on a statistical basis. The food balance sheet can never provide information on the diets of individuals. Nor does the family dietary survey as usually conducted, although the diets of individuals can be measured if specially designed surveys are made.

III. STRONTIUM

Emphasis is given to consideration of Sr-90 since this nuclide represents the long-term hazard, although the behavior described would also be valid for Sr-89.

Behavior in soils and plants

The evidence now available for 1958 shows that the Sr-90 from world-wide fallout which has entered the diet of livestock and man is often derived largely from recently deposited material. Surface contamination of vegetation and foliar and plant-base absorption appear particularly important in pastures; floral absorption may contribute much Sr-90 to cereals. It is impossible to estimate precisely the contribution of absorption from the soil to the total contamination since considerable variations occur among soils, crops, meteorological conditions and the patterns of fallout. In some regions it appears that in 1958 the absorption from the soil contributed only about one fifth of the Sr-90 in milk; quantitatively different relationships can be expected in different regions.

Thus, where a major part of the plant contamination has been derived from recent deposition of Sr-90, a decreased rate of fallout may be expected to cause a decrease in the Sr-90 content of plants even though the cumulative deposit were to rise. The contribution of absorption from soil relative to direct contamination will, however, increase and the Sr-90 content of produce which is particularly subject to direct plant contamination (milk and cereals) should decline relative to other foods. Even when only a small part of the Sr-90 in plants has hitherto entered from soil, the Ca content of soil may nonetheless be a significant factor affecting the ratio of Sr-90 to Ca in diet. A low concentration of Ca in the soil will not only reduce the concentration of Ca in plants, but it will also enhance the absorption of Sr-90 from the soil; both effects will increase the ratio of Sr-90/Ca in plants.

Animals and animal products

While the concentration of Sr-90 in a tissue, secretion or food is the essential information for evaluation of the Sr-90 hazard, it appears that consideration of strontium-calcium ratios is of much greater

usefulness than consideration of strontium retention in animal or man. It is emphasized, however, that workers should report survey values of Sr-90 in terms of both calcium content and weight of product.

The differential behavior of strontium and calcium has been expressed as the Strontium-Calcium Observed Ratio (OR) and defined as follows:

$$\text{OR sample/precursor} = \frac{\text{Sr/Ca of sample}}{\text{Sr/Ca of precursor}}$$

OR body/diet values for laboratory and domestic animals under usual dietary conditions appear to range from 0.18 to 0.35 with most of the values falling around 0.25. A preponderance of data in the United Kingdom indicate an OR body/diet value for man of 0.25; however, other limited studies have indicated a range up to about 0.5. The OR milk/diet for domestic animals appears to range from 0.09 to 0.16 with most values falling close to 0.11.

The concept of the Observed Ratio may permit:

- (a) prediction of total body burdens from dietary values;
- (b) prediction of total diet values from tissue or excretion measurements;
- (c) prediction of maximum Sr-90/calcium ratios of any single mineral deposit in the body from measurements on diet.

Aquatic food chains

From limited data it appears that in 1958 there was about 1 μc /liter of Sr-90 in the surface water of the Pacific Ocean, to a depth of about 100 meters; other oceans had about one tenth of this value. Fish from the Pacific Ocean in 1958 had about 0.3 μc Sr-90/gram Ca. It is emphasized that human consumption of even small fish including bones would cause only minor contribution to the Sr-90 burden of the consuming public.

The distribution of Sr-90 in freshwater environments is much more variable than in the marine environment, and in general fresh-

water fish have higher Sr-90 to calcium ratios than marine fish by as much as twentyfold. However, the contribution of Sr-90 to the human diet from freshwater fisheries products also remains small compared to the contribution from land sources.

Diet composition

In countries where relatively large amounts of dairy produce are used, these items have been the single most important contributor of Sr-90 to the human diet, with grain next in importance, and other components making small individual contributions; water and fish are of minor importance. Further data are needed to evaluate dietary Sr-90 contributions for specific dietary and regional situations. The resultant dose of radiation to the skeleton is closely related to the ratio of Sr-90 to calcium in the total diet.

As the Sr-90 content of diet is considered in terms of the content of calcium, the first step is to review the dietary sources of calcium of the population concerned. These vary widely in different countries.

Study of data from food balance sheets suggests that those countries for which information is available can be grouped into four broad categories: *the first*, which includes most of Europe, North America and Oceania, with food supplies containing over 800 milligrams of calcium per head per day, of which 70 to 90 percent is obtained from milk and milk products; *the second*, which includes some Southern European countries, such as Italy, and some Latin American countries, such as Argentina and Uruguay, with 600 to 700 milligrams of calcium daily, about 60 to 80 percent of which comes from milk and milk products, but in which countries vegetables tend to make a relatively more important contribution than in the first group; *the third*, which includes Egypt, Turkey, India, Pakistan and the Union of South Africa, in which milk and its products are also the main contributors (30 to 65 percent), but to a much smaller total calcium content of 300 to 450 milligrams, and in which cereals, pulses and nuts, and vegetables become relatively more important; and *the fourth*, which includes Chile, Ceylon and Japan, where milk and its products do not make the major calcium contribution and where

the daily consumption of 200-350 milligrams per head is derived fairly evenly from cereals, pulses and nuts, vegetables and dairy produce. In Chile and Japan fish also makes an important contribution. Few data are available for Africa and other undeveloped areas of the world.

Similar impressions can be gained from a study of the results of the small number of family food consumption surveys that have been made. From such surveys it is usually possible to derive information on unusual sources of calcium of mineral as well as animal and vegetable origin and to learn something of the forms in which foods are actually eaten, that is after they have been subjected to processing and domestic preparation.

Illustrative survey levels in diets

Surveys have been conducted in a systematic manner in Japan, the United Kingdom and the United States. The results of these surveys are summarized in Appendix V.

The ratio of Sr-90 to calcium in the United Kingdom diet has been calculated as about $6 \mu\mu\text{c Sr-90/g Ca}$ in 1958. Estimates for the United States for the same year appear to be about double that value. Japanese values for 1959 ranged from 11 to 15 $\mu\mu\text{c Sr-90/g Ca}$ in different parts of the country.

IV. CESIUM

Behavior in soils and plants

In contrast with Sr-90, Cs-137 is usually retained in the soil in forms which make it largely inaccessible to plants. The contamination of plants is, therefore, primarily due to direct deposition. The movement of Cs-137 in plants also contrasts with that of Sr-90; Cs-137 moves freely throughout the plant whereas Sr-90 is not transferred from leaves to other tissues to an appreciable extent. Direct contamination can, therefore, lead to higher concentrations of Cs-137 than Sr-90 in tubers.

Animals and animal products

Radioactive cesium is efficiently absorbed from the gastro-intestinal tract, secreted into milk, and accumulates within the cells of the body, being physiologically similar to potassium. An important feature is the relatively rapid turnover of cesium within the body, which varies with body size from a few days in laboratory animals to about 20 days in cows. The behavior of cesium, however, appears to be independent of potassium levels.

Aquatic food chains

From limited data it appears that the Cs-137 concentration in fish from the Pacific Ocean in the period 1954-1958 was not more than about $0.02 \mu\mu\text{c}/\text{gram}$ of edible fish. The concentration in fish from the other oceans would be somewhat lower, and from fresh water would be somewhat higher. In any event, fisheries produce could only contribute a very small fraction of the Cs-137 in the human diet.

Diet composition

Analyses for Cs-137 already made in Canada, the Federal Republic of Germany, Norway, Sweden, the United Kingdom and the United States of America lead to the view that the most important dietary sources of this isotope are milk and meat. Available data are, however, very limited and no general statements on the effect of diet composition on the intake of Cs-137 can be made until the subject has been more fully examined. There is some evidence that, in countries in which the potato is an important article of diet, it may also be a significant contributor of Cs-137. No data are available for countries whose food supplies contain large proportions of cereals or starchy roots other than potatoes.

Illustrative survey levels in diet

Calculations have been made which suggest that in the United States of America about 60 percent of Cs-137 in the human diet comes from milk, 25 percent from meat, 7 percent from flour and

cereals, 5 percent from vegetables and 3 percent from citrus fruits. A study in the United Kingdom suggests that, for certain individuals, milk contributed about one half of the Cs-137 content of the diet; meat, fruit and potatoes were the other important sources. Data from Germany also indicate that the potato may be a relatively important source.

Data for 1957-58 from several countries in the Northern Hemisphere showed a range of 20 to 90 $\mu\mu\text{c}$ of Cs-137/gram of potassium in milk. A few samples from Argentina gave an average of about 9 $\mu\mu\text{c}$ of Cs-137/gram of potassium.

V. IODINE

Because of the short half-lives of the significant radioactive isotopes of iodine it is only necessary to consider the contamination of foods which may be consumed by man shortly after contamination has occurred. Milk is the outstanding example.

The quantity of I-131 which reaches milk will depend on the extent to which the deposit is intercepted on plants which cattle consume. The extent of this deposit will vary greatly because, among other matters, of variations in the nature of the herbage and because I-131 may be removed from leaves in rain to an appreciable extent.

On the average, it is indicated that 5 to 10 percent of the ingested I-131 appears in the milk of the dairy cow and 20 to 30 percent in the milk of the goat and sheep. There seems to be considerable variation between individuals possibly because of such factors as season, feeding practices, stage of lactation and status of the thyroid gland.

VI. OTHER RADIOACTIVE SUBSTANCES

Radioactive nuclides that are poorly absorbed from the gastrointestinal tract of man and animals and those which do not reach the edible products of animals are of minor concern in terrestrial food chains. Brief mention is made of the metabolism in domestic animals of other possibly important radionuclides. Limited data

indicate that the following percentages of ingested materials appear in the milk of the cow: Ba-140, 0.4 percent; zinc, 5 percent; cobalt, 0.1 percent, iron, 0.1 percent. The dose contribution of these radionuclides to the human diet is at present small compared to that of the biologically important fission products.

In regard to C-14 it is pointed out that the specific activity in any part of the food chain would probably lag by less than a year that of the C-14 used by plants for photosynthesis.

Because some marine organisms have high concentration factors for certain fission products, such as Ce-144, and for certain induced radionuclides, such as Zn-65, Fe-55 and Fe-59, and Co-60, these substances are of greater importance in the sea than on land. Measurement of the Ce-144 concentration of the surface waters of the Atlantic Ocean in 1958 gave values of the order of $0.5 \mu\mu\text{c/liter}$. Concentration factors of edible marine products of the order of 10 to 100 have been found for this element, so that a concentration of Ce-144 of $0.05 \mu\mu\text{c/grams}$ of edible fish would be the maximum which it is likely now occurs. A high daily intake of 100 grams of sea food would therefore lead to a daily intake of Ce-144 of $5 \mu\mu\text{c}$ per individual. Even this maximum estimate constitutes a small contribution compared to the total daily intake of Sr-90 from all sources, but does approximate the contribution due to Sr-90 from marine products alone. However, the relative importance of this radionuclide will decrease with time because of the shorter half-life compared to Sr-90.

Measurements of Zn-65 and Fe-55 in fisheries products from the Pacific indicate that a population having the relatively high intake rate of 100 grams of marine products per person per day would ingest about $1 \mu\mu\text{c}$ of Zn-65 and $10 \mu\mu\text{c}$ of Fe-55 per person per day. Products from other ocean areas would be even less contaminated. These isotopes then contribute much less radiation dosage than Sr-90 in sea food products, which has already been shown to constitute only a small fraction of the total daily intake.

Isotopes which are highly concentrated by marine and fresh water organisms are likely to be components of waste materials from peaceful uses of nuclear energy introduced into the sea. While such introductions are unlikely to be significant insofar as large population groups are concerned, they may become important in limited areas for limited population groups.

VII. REMEDIAL MEASURES

GENERAL

It is possible that the increasing use of nuclear energy could lead to serious contamination of the food chain with radioactive materials which may require remedial measures of effectiveness beyond that of natural processes or of normal management and manufacturing practices. Reduction of contamination of successive stages of the food chain could result in an over-all degree of decontamination equivalent to the mathematical product of the respective individual decontamination factors. Investigations of a number of approaches to the problem are under way. Expansion of research in this field is needed, so that we may be prepared to deal adequately with situations that may arise in the future.

Any emergency measures should be applied only when they are declared necessary by the responsible authorities. Decisions as to choice of measure would have to be based on the anticipated effectiveness of a method, together with such factors as attendant reduction in food supplies, expense, and disruption of economic and dietary patterns.

SOILS AND PLANTS

Remedial treatments should be applied to land only under the guidance of soil and crops advisors, and only when declared necessary by the responsible authorities.

Experiments indicate that, although the removal of recently contaminated annual crops generally would not be a highly effective land decontamination measure, removal of grass sod could be expected to eliminate over 90 percent of the contamination. Scraping of several inches of surface soil by a small road grader has had variable results, but decontamination of nearly 100 percent effectiveness was achieved when loam soil had a smooth seedbed surface. Preliminary small-scale experiments have indicated potentially high effectiveness of spraying an asphalt emulsion onto smooth contaminated soil and the subsequent removal of the hardened crust.

Leaching with large quantities of acids or soluble calcium salts

has been found experimentally to move strontium to a considerable depth, but soil fertility or structure would probably be damaged thereby. Field experiments indicate that, where soil conditions permit and the necessary equipment is available, deep plowing can be of some effectiveness for shallow-rooted grasses.

Where liming of acid soils is needed for optimal crop productivity, it often also reduces the strontium content or ratio of strontium to calcium in the crop. However, additions of lime beyond the requirement for this purpose would be of no practical value in reducing the hazard, and extremely large reduction factors should not be expected from this practice.

ANIMALS

Livestock, together with uncontaminated feed and water supplies, can be placed in shelter in order to avoid their ingestion of radionuclides. Where animals must be fed on contaminated pasture, their foraging should be restricted to the minimal area which can provide adequate maintenance. Experimental work with cows and goats has indicated that under proper conditions additions of uncontaminated calcium to the food may effect reductions of about two to three fold in the ratio of Sr-90 to calcium in the animal's bone or milk. Before such methods could be applied, careful physiological and nutritional studies would have to be done especially if there were to be consideration of alteration of human diets.

CHANGES IN FARMING SYSTEMS

When land is seriously contaminated with Sr-90, changes in farming systems could be considered as a means of providing food supplies acceptable from the viewpoint of radiation hazard to humans. The selection of alternative crop and animal production systems would be based on the reduction of the content of soil-derived Sr-90 in the diet, primarily that of the younger people in the population. The pertinent agricultural, economic, and nutritional problems accompanying such a change would have to be given adequate attention in each case.

ANIMAL PRODUCE

Laboratory experiments have indicated that about 85 percent of the radioactive strontium and radioactive cesium in cow's milk can be removed by treatment with cation exchange resins saturated with nonradioactive cations, without apparent lowering of the quality of the milk. Radioactive iodine has been removed from milk up to the extent of 98 percent by treatment with an anion exchange resin in the chloride form. Neither of these methods has been studied yet on a pilot plant scale. Preliminary laboratory experiments have been conducted on the removal of Cs-137 from meat by treatment with sodium chloride.

VEGETABLE PRODUCE

External contamination of raw fruits and vegetables can be reduced by various surface treatments. These might include a scrubbing or washing treatment followed by the appropriate skinning procedure. Where the whole grain or its products of wheat, rice and other grains normally is eaten, discarding the bran and outer layers would eliminate a substantial fraction of the Sr-90 of the grain from the diet, as Sr-90 is relatively concentrated in the bran. Since this would also reduce calcium, vitamin B₁ and other nutrients in the diet, they might have to be supplied from other sources.

VIII. SURVEYS OF ENVIRONMENTAL RADIONUCLIDES IN FOOD AND AGRICULTURE

It is desirable that any survey of the levels of radionuclides should be undertaken with a clear definition of the objective and an assessment of the degree of accuracy of measurement required to achieve the objective. In deciding upon the objective, the use of the survey for assessing the possible hazards to man should be given special consideration. Such an examination may show that facilities are not available for a survey of the required accuracy, or that the survey may utilize available technical personnel and equipment to an extent not justified by the objective. Where the objective is recognized as of undoubted importance to a large area or popu-

lation, governments should undertake the work to the extent feasible and international support should be provided where necessary. It may also be desirable to consider organizing certain aspects of the work on a regional basis.

A survey program may have more than one objective. Where this is so it is essential to decide on the relative importance of the objectives and to avoid compromising the important in the hope of obtaining some information on the less important aims.

Much of the information obtained from such surveys will be of value in estimating the possible hazard to man.

Examples of the objectives of surveys of the levels of radionuclides are:

- (a) determination of concentrations of radionuclides in key food and agricultural materials on a local, national or regional basis;
- (b) the estimation of the level of specific radionuclides in a human population;
- (c) the estimation of the levels of radionuclides in the diet of a human population, with, where possible, a breakdown by principal items of diet;
- (d) the use of the information obtained under (b) and (c) as a factor in predicting future trends in radionuclide concentrations.

In all surveys it is essential to agree in advance the degree of accuracy required. If possible, the survey should also give a statistical distribution and values of mean and a standard error for the mean or a similar computation.

Much more information on the behavior of radionuclides in the environment is needed. This will be obtained partly from field and laboratory experiments and partly from direct studies of the physical and biological environment, e.g., studies in ecology, agriculture and nutrition. Environmental surveys have a definite place in the over-all program of research.

Certain further general principles in the design of surveys can be stated.

- (a) A survey dealing with environmental radioactive contamination cannot be rationally designed without a clear understanding of the basis on which biological effects are to be estimated, e.g., whether the effects are governed by a threshold value.

Unfortunately, medical knowledge of the effects of low radiation levels is still subject to major uncertainties. The planning and interpretation of radionuclide surveys will be advanced to the extent that these uncertainties are diminished.

- (b) Where levels of radionuclides are to be measured in a complete diet or individual agricultural products, both sampling and analytical techniques are involved. A sample for subsequent analysis has to be drawn from a bulk made up from a number of subsamples taken at different points and over an adequate period of time.

In order to determine the size of the final sample, it is necessary to estimate the order of magnitude of the level of activity to be expected.

The sampling must be so designed that the final sample for analysis is truly representative of the material under examination. The general methods and statistical techniques for achieving this are well-established, and advice on them should be available to any regional or national body designing a survey.

- (c) In large programs involving repetitive routine radiochemical analysis it is essential to maintain a continuous control on the standards of accuracy of analysis and measurement and to provide for proper interlaboratory comparisons. This involves the services of well-trained scientists; the necessity for this control is enhanced where sampling and analysis are separated.
- (d) Although methods of radiochemical analysis have been worked out with a considerable degree of refinement, there is still ample scope for their improvement and particularly the reduction of the labor involved, and thereby to increase the scope and accuracy of environmental surveys.
- (e) Co-ordination of effort at national, regional and international levels is of the greatest importance. The value of surveys carried out in different areas will be greatly increased if they are designed so that the results may be integrated. It is equally important that surveys carried out for different purposes within one country or region should be co-ordinated. Thus meteorological, geophysical, ecological, medical, agricultural and dietary surveys can be of great value in designing and interpreting surveys of radionuclide levels if this purpose has been kept in mind in co-ordinated planning.

While the value of environmental surveys of the levels of radionuclides is fully recognized, *it is emphasized that it is important to examine continuously the value of this type of work in relation to the effort involved.*

IX. RECOMMENDATIONS

The Committee recommended:

1. that research and development of concepts on the behavior of all possibly important radionuclides in the terrestrial and aquatic food chains should be encouraged;
2. that detailed studies of the behavior of fission products, especially Sr-90 and Cs-137, in plants and soils be conducted to provide information on the mechanisms which control the contamination of plants: the aspects on which information is particularly lacking are:
 - (a) knowledge of the redistribution of fission products after deposition and of their behavior over long periods in exchange systems and fixation processes in the soil,
 - (b) knowledge of the effect of climate, plant form, soil type and cropping systems on the relative extent to which Sr-90 and Cs-137 enter plants directly or through the soil,
3. that research should be also undertaken on other radioactive substances which, because of their long half-lives or possible entry into biological systems, could be a source of concern under particular circumstances: this category includes isotopes of iodine and plutonium;
4. that the study of the behavior of naturally occurring radioactive substances, especially the radium and thorium series, in soils, plants and agricultural products should be encouraged;
5. that in view of the fact that, at least for limited areas and limited population groups, radioactive materials originating from peaceful uses of nuclear energy introduced into freshwater and marine environments may become important, it is recommended that research be conducted on the movement through the

aquatic food chains of these radionuclides which are likely to become important in such introductions;

6. that studies of remedial measures including research on agricultural processes and on decontamination of food products should be continued to include all dietary items that are relatively important carriers of radionuclides;
7. that up-to-date information on the organization of national environmental radionuclide survey programs relating to food and agricultural materials, including objective and scope, framework of data interpretation, techniques of sampling and sample analysis, types of radionuclide measurements, specific nuclides measured, plans for reporting data and results, and plans for relating to similar programs elsewhere, be collected and distributed on a continuing basis;
8. that, since the objectives of surveys will differ from country to country depending on internal factors including diet and physical environment, and the nature of food and agricultural samples to be taken will depend on the objectives, detailed decisions on such matters should be based only on expert consideration of conditions in the country or region involved; in this connection the interested agencies of the United Nations could be of assistance by providing short-term technical advisors to governments where necessary;
9. that, since close co-operation between various scientific disciplines such as chemistry, physics, biology, agriculture, and medicine is essential in the organization of surveys, all such programs, both on a national and international basis, should be under the regular surveillance of representatives of these disciplines;
10. that individual nations and the interested agencies of the United Nations give consideration to providing the following facilities to assist member nations in food and agricultural survey work:
 - (a) expert advice in the design of surveys where this is required, including the necessary statistical services,
 - (b) advice and assistance in the standardization, interpretation and reporting of results,

- (c) training in sampling methods, i.e., in the detailed methods for taking, handling and transporting samples.
 - (d) a radiochemical analytical service available to nations which lack the necessary staff or facilities,
 - (e) training in radiochemical techniques,
 - (f) assistance to individual nations desiring to co-ordinate survey programs with other nations;
11. that the study of methods of obtaining reliable estimates of food consumption in sufficient detail should be strengthened, particularly in underdeveloped countries: as the composition of the diet has a controlling influence on the intake of nuclides, it is important to obtain adequate information, especially on the lesser known diets of the world: reliable estimates of food consumption should be used as the basis for monitoring programs and for calculation of the content of nuclides in diet;
 12. that, since it is easier to obtain records of the average food consumption of a population than that of any particular group, such as young children, on whom it may be necessary to obtain dietary information, it is recommended that methods of obtaining satisfactory information on the food consumption of such groups be studied: other types of surveys, e.g., agriculture, food consumption and nutrition, can provide supplementary information of great value;
 13. that, as the composition of the total diet determines the effective internal dose of radiation to the body, it is important to relate analyses of individual foods to their contribution to the diet as a whole: absolute levels of nuclides and nutrients should be reported so that ratios can be calculated as required;
 14. that, since food represents the major pathway for transmission of environmental radioactive nuclides to man, and because of the specialized knowledge of many food, agricultural and fisheries experts, governments make provision for direct technical representation by food, agricultural and fisheries authorities on appropriate national or international radioactivity committees, or for the establishment of food, agricultural and fisheries committees or subcommittees to act in an advisory capacity to such over-all radioactivity committees;

15. that adequate consideration of the local food-producing environment, including agricultural conditions and practices, soils, underground, surface and marine waters, around the projected sites of nuclear energy installations, given in close consultation with food, agricultural and fisheries authorities, should be regarded as an essential part of the planning for such installations and of the subsequent radiological control needed during operation of the installation;
16. that, in order to keep radioactivity in food, agriculture and fisheries under continuing review on an international basis, and to stimulate research in this field, the Director-General of FAO should convene periodically technical expert committees on this subject, on either an over-all or a limited subject-matter basis, as may be most appropriate in the prevailing circumstances, and with membership adequately representative of differences in regional environmental and agricultural conditions and dietary customs that should be taken into consideration: one such group should in the near future consider and advise on food survey and sampling techniques and on the methods of assessment of results; another group should consider potential fisheries problems associated with the peaceful uses of atomic energy;
17. that, in view of the need for disseminating information on radioactivity in food, agriculture and fisheries, encouraging research in this subject under an adequately wide range of conditions, and providing opportunity for personal contacts between experienced research workers and those who are newly developing research programs in this field, the Director-General of FAO should convene, from time to time, technical meetings or symposia, on either a comprehensive or a restricted disciplinary basis as may be most appropriate;
18. that, because of the rapidly developing use of atomic energy for a variety of purposes and the increased levels of environmental radioactivity that can arise therefrom, with the consequent need that FAO be in a position to advise and assist the food, agricultural and fisheries authorities of its Member Governments in their responsibilities for research on, and the control of, environmental radioactivity in relation to food, agriculture and fisheries, increased emphasis be placed on work in this field in the program of work of the Organization.

RADIOACTIVE NUCLIDES OF IMPORTANCE IN RELATION TO POTENTIAL CONTAMINATION OF HUMAN FOOD SUPPLIES

The various applications of atomic energy involve the production of very large quantities of radioactive fission-product nuclides presenting a potential problem in regard to the contamination of human food. Apart from the fission products released to the environment in nuclear explosions, large quantities are produced in nuclear reactors and are held largely as waste materials following the processing of the fuel in chemical separation plants. On account of the release of neutrons in the fission process, materials exposed to high fluxes of these neutrons become activated and some of the resulting nuclides may be of biological significance if they are released into the environment. In addition to the artificial radioactive nuclides, there are several naturally occurring radioactive elements notably the long-lived, mostly alpha-active, isotopes of the uranium, thorium and actinium series and also the element potassium of which the K-40 isotope exhibits beta-activity. These materials enter the human food chain and are present in the human body. The purpose of the present appendix is to present a broad general view of the relative importance of the various nuclides, but it must be recognized that, in special circumstances, individual isotopes not mentioned specifically could also be of biological concern.

Radionuclides produced in fission

The atoms produced in the fission process fall mainly into two groups with atomic weights between 80 and 108, and 126 to 154. Between these groups the fission yield is comparatively low. The yield curve is roughly the same for the different fission materials U-235, U-233, and Pu-239, although for the fast-neutron fission

(including that of U-238) there is some increase of yield in the trough between the two main groups. The most prominent fission products are produced with a yield of about 6 percent of all fissions. Table 1 shows the fission products for which the yield is greater than 0.03 percent of all fissions and where the half-life is greater than 10 hours and less than 5×10^5 years (1).¹ The radioactivity of any nuclide can be calculated from the fission yield given in Table 1 since the disintegration constant (sec^{-1}) is equal to 0.693 divided by the half-life in seconds. There is no necessity to include isotopes of very long half-life since their specific radioactivity is low. An example of this is the iodine isotope, I-129, which, although it is produced in about 1 percent of all fissions, has a half-life of 17 million years; as a consequence, the specific radioactivity of the isotope would not be sufficient to distinguish it from ordinary stable iodine in regard to its biological effects.

The fission products are formed in chains in which the nuclides are of the same mass number, the transmutation from one nuclide to the next occurring as a result of radioactive beta-decay. The relative yields for the various chains shown in Table 1 would be applicable to an event in which all the fissions occurred at substantially the same time such as in a nuclear explosion or a criticality accident (although in the latter case the amount of radioactivity produced would be relatively very small). The proportions would also be applicable in the event of a power excursion in a reactor which had only recently been started after a change of fuel.

Fission products accumulated in a nuclear reactor

When a nuclear reactor is operated for a considerable time the long-lived activities build up progressively, whereas for the short-lived activities the rate of decay rapidly becomes equal to the rate of production and an equilibrium level of radioactivity is reached. In view of this the proportions of the various isotopes change progressively with time. A somewhat similar situation arises in regard to the build up of activity when nuclear explosions are conducted on a continuing program. Table 2 shows the amounts expressed

¹ Numbers in parentheses refer to authorities quoted under "References" at the end of this Appendix.

TABLE I. - FISSION PRODUCTS IMPORTANT ON ACCOUNT OF YIELD
AND HALF-LIFE

(Atom yield > 0.03 percent; half-life > 10 hours)

Chain atomic weight	Fission yield atom percent	Nuclide and half-life
85	1.3	Kr (10y)
89	4.6	Sr (51d)
90	5.1	Sr (28y) - Y (61h)
91	5.4	Sr (9.7h) - Y (57d)
93	6.0	Y (10h)
95	6.3	Zr (65d) - Nb (35d)
97	6.4	Zr (17h) - Nb (74m)
99	6.0	Mo (68h) - Tc (5.9h)
103	3.4	Ru (40d) - Rh (57m)
105	1.0	Ru (4.5h) - Rh (36h)
106	0.5	Ru (1.0y) - Rh (30s)
127	0.16	Sb (93h) - Te (90d 20% 9.3H 80%)
129	0.9	Te (32d, 21%)
131	3.1	I (8.1d)
132	4.0	Te (78h) - I (2.4h)
133	6.3	I (22h) - Xe (5.3d)
135	6.0	I (6.7h) - Xe (9.2h)
137	6.2	Cs (37y) - Ba (2.6m)
140	6.1	Ba (12.8d) - La (40h)
141	6.0	Ce (33d)
143	5.0	Ce (33h) - Pr (13.7d)
144	5.0	Ce (290d) - Pr (17.5m)
147	2.9	Nd (11.6d) - Pm (3.7y)
149	1.4	Pm (47h)
151	0.5	Pm (27.5h) - Sm (73y)
153	0.16	Sm (47h)
155	0.03	Eu (1.7y)

TABLE 2. - FISSION PRODUCTS IN NUCLEAR REACTOR
(Running 1 year; cooling 1 day)

Total activity about 820 kilocuries/mw			
Chain atomic wt.	Isotope and half-life	Fission yield %	Activity* kilocuries per megawatt
89	Sr (53d)	4.6	39
90	Sr (28y)	5.1	1
91	Y (57d)	5.4	45
95	Zr (65d)	6.3	53
103	Ru (40d)	3.4	28
106	Ru (1y)	0.5	2
131	I (8.1d)	3.1	24
132	Te (78h)	4.0	30
133	I (22h)	6.3	24
137	Cs (37y)	6.2	1
140	Ba (12.8d)	6.1	48
141	Ce (33d)	6.0	51
144	Ce (290d)	5.0	26

* Excludes daughters

as kilocuries per megawatt of power of some representative isotopes in a nuclear reactor which has been running for one year. While those proportions are quoted for 24 hours after the shutdown, the proportions at any subsequent time would depend upon the relative rates of decay of the various nuclides. The proportions which are relevant to environmental contamination depend, however, on a number of other important factors which are considered below.

Factors influencing possibility of escape of radioactivity in a nuclear reactor accident

In most nuclear reactors the fuel in which the nuclear fission takes place is in the form of metallic bars enclosed in hermetically sealed cans. In many instances these are arranged in channels in a

graphite moderator. In other designs they are supported in water or heavy water, which may be under very high pressure in a hermetically sealed enclosure. In some instances the nuclear fuel is supported in channels through which liquid sodium is pumped under pressure. In virtually all power reactors, the reacting core, or probably the whole reactor, is enclosed in a hermetically sealed pressure vessel designed to contain the contents in the event of any engineering failure.

The possibility of the significant escape of radioactivity from nuclear power reactors, which are at present envisaged, is thus very small. Moreover, the existing experience in the nuclear reactor accident with an early type of open reactor at Windscale in 1957 showed that isotopes of the volatile elements iodine and cesium and, to some extent, the ruthenium and tellurium, escaped from the reactor whereas the strontium and cerium were retained to the extent of something like 99 percent in the fuel or graphite moderator of the reactor. These considerations suggest that the isotopes most likely to escape are those of the volatile elements, especially iodine and cesium and, of course, the noble gases which would escape whenever the integrity of the fuel and the enclosure is breached, but it is prudent to include strontium for consideration on account of its biological importance, despite the smallness of the fraction likely to be released.

From the very nature of accidents it is not possible to predict precisely what is likely to occur. However, some indication of the possible extent of contamination from the escape of a given quantity of radioactivity can be judged from the Windscale experience. Fairly extensive surveys of the conditions in the environment have already been published (3, 4, 5, 6). The estimated release of radio-iodine I-131 at Windscale was 20,000 curies from a high stack. Milk contamination was such that in areas where the herbage level was 1 μc square meter the milk level was about 0.1 μc /liter. Relatively higher milk contamination could arise in summer conditions but in winter milk contamination would be likely to be small. The I-131 contamination of other human foodstuffs was by comparison small. In contrast with iodine, contamination from isotopes of strontium (3, 7, 8) did not necessitate restriction of consumption of food or milk. Criteria for acceptable levels for contamination of dietary items applicable in an emergency have been evaluated

in the United Kingdom (9). In this connection also reference should be made to the report of the joint WHO/FAO Expert Committee on Methods of Radiochemical Analysis (15).

The extent of the hazard in regard to nuclear power plants depends upon the extent to which fission products can be certainly contained in any eventuality and on the corresponding estimation of the maximum credible accident (2). In view of the developments in reactor engineering and, in particular, in the design of reactor enclosures, it is exceedingly unlikely that more than a small fraction of the fission product content of a reactor could escape in an emergency with the types of advanced power reactors now being designed.

Biological factors affecting hazard from ingested fission products

The biological properties of the various nuclides greatly affect the relative importance. From Tables 1 and 2 it will be seen that the important fission product nuclides are those which comprise the rare earths, the zirconium-niobium isotopes, the noble metals especially ruthenium and rhodium, and the alkaline earths especially strontium and barium. Some indication of their relative importance biologically can be seen by inspection of the fractional uptake from the gut of these various groups of isotopes and this is shown in Table 3. It will be seen that although the rare earths comprise an important group of fission products, the uptake from the gastro-intestinal tract is so small both in man and animals that, although they irradiate the gastro-intestinal tract in passage, they are of little importance in the food chain. Similarly, zirconium-niobium are taken up to only a slight extent and we are left with the alkaline earths, strontium and barium, the alkali metal cesium, along with tellurium and the iodine isotopes as of predominant importance. It appears unlikely that any other fission product isotope would be anything like as troublesome in a food chain involving mammals, such as the cow, and uptake through the human gastro-intestinal tract. In considering escape of fission products into water, however, the situation is modified by the likelihood of some of the radioactivity, particularly of rare earth elements, being concentrated in certain marine organisms.

TABLE 3. - FISSION PRODUCTS OF BIOLOGICAL IMPORTANCE

Chemical character	Isotopes important on account of yield and half-life	G Uptake from G.I. tract to:	
		Total body	Critical organ
Halogens	I ¹³¹ I ¹³³ I ¹³⁵	1.0	0.3 thyroid
Oxygenated anions	Te ¹³² -I ¹³²	0.25	
Alkali metals	Cs ¹³⁷ -Ba ¹³⁷	1.0	0.4 muscle
Alkaline earths	Sr ⁸⁹ Sr ⁹⁰ -Y ⁹⁰	0.3	0.2 bone
	Ba ¹⁴⁰ -La ¹⁴⁰	0.05	0.04 bone
Rare earths	Y ⁹¹ Zr ⁹⁵ -Nb ⁹⁵	10 ⁻⁴	3 × 10 ⁻⁵ bone, liver
	Ce ¹⁴¹ Ce ¹⁴⁴ -Pr ¹⁴⁴ Pr ¹⁴³ Nd ¹⁴⁷ Pm ¹⁴⁷		
Noble metals	Ru ¹⁰³ Ru ¹⁰⁶ -Rh ¹⁰⁶	0.03	4 × 10 ⁻³ bone, kidney

¹ Isotope pairs are classed according to chemical and biological character of parent isotope.

² The uptake of Pu from G.I. tract is 3 × 10⁻⁴.

Nuclides produced by activation

In addition to the production of fission products, the fission process leads to the activation by neutrons of materials in the vicinity. In atomic explosions, activation products include isotopes of iron and zinc from the activation of structural materials; these and other activities may also be produced in sea-water. The most important activation product is the radioactive isotope C-14 arising from the reaction of neutrons with nitrogen atoms in the atmosphere. The level of activity of this isotope in the troposphere has been reported as 10 to 15 percent above the natural level as of mid-1958. If there are no more nuclear explosions the average level in the atmosphere is expected to fall with the progressive exchange with the rest of the carbon reservoir. Further residual products of biological concern from nuclear explosions include plutonium and tritium.

Activation products also arise in the operation of nuclear reactors and may in some instances escape to the environment. For instance, in reactors using closed circuit carbon dioxide gas as a heat-transfer

material C-14 is produced by activation of the stable C-13 and the transmutation of traces of nitrogen in the carbon dioxide. This isotope has a very long half-life and consequently exhibits very small specific radioactivity. On account of the role of carbon in biological processes, C-14 enters into the metabolism of plants and into the food chain, but the dilution is usually so great that the C-14 produced by reactors is of no biological significance even when the whole carbondioxide content of a nuclear reactor is discharged.

Neutron activated isotopes are important where water, used as a heat-transfer material in reactors, is discharged to a river system and there is extensive published information on the fate, particularly of a large number of isotopes produced in the cooling water (10). In nuclear power reactors the coolant is usually operated on a closed circuit and environmental contamination only arises from the periodic discharge of coolant for purposes of fuel changing or maintenance of the reactor or from slight leakages which might occur from time to time. The activation of nuclides in reactors with the possibility of discharge especially as liquid wastes involves particularly the following isotopes: Co-60, Fe-55, Fe-59, Cr-51, Cu-64, Ta-182, Zn-65. It may be difficult to avoid disposal of small quantities of these isotopes in the sea from nuclear-propelled ships (14). Many other isotopes may be discharged in small quantities in specialized types of plant, such as P-32 and Na-24 to the Columbia River at Hanford (10). With sodium-cooled reactors there is also the possibility of the accidental escape of activated sodium into the environment or its reaction with water used as a moderator, although it is not very difficult to prevent hazards of this kind by appropriate engineering provisions.

Chemical separation plants

The fuel used in nuclear reactors must periodically be chemically processed in order to remove fission products which interfere with the functioning of the reactor if allowed to accumulate indefinitely, and on account of metallurgical changes in the fuel. The fission products are separated by chemical processes from the uranium or plutonium which may then be refabricated into fuel elements.

When the nuclear fuel is dissolved certain of the fission product isotopes can readily become airborne including, of course, all the rare gas isotopes. It is difficult but not impossible to trap these and to pump them into storage bottles, but this procedure is not at present

justified and it should be noted that there are no food chain problems from the rare gases. Residual amounts of the radio-active iodine I-131 may also be released to the environment but the amount is usually controlled by ensuring an adequate storage period for the decay of this isotope and it can furthermore be removed by chemical procedures.

The amount of radioactivity produced in chemical separation plants is very large and represents the long-lived fission products which have accumulated with very little radioactive decay during the period of operation of the reactors. It has been customary hitherto to store these long-lived activities in a highly concentrated solution in durable tanks (11). The location of these is usually such that escape into the sub-soil would not produce any contamination of human drinking water or farmland and, in any case, the tanks are usually arranged so that leakage can be detected immediately. Methods have now been developed, moreover, by means of which it is possible to convert the highly concentrated high-activity wastes into the form of a durable solid material such as glass which can then be stored safely (12).

In the course of operation of the chemical separation plants it is usually necessary to devise some method of dealing with dilute radioactive wastes which are so voluminous that they cannot be stored. There are also economic difficulties in concentrating such activities and, in many instances, it is more economic to dilute down to a safe level and discharge these rather than to attempt to accumulate them on the site. By means of well-known chemical flocculant precipitation methods most of the heavy elements including fissile material and the alpha activities can be largely removed. Difficulties arise, however, in connection with certain beta-active isotopes and small amounts of these, such as ruthenium, must usually inevitably be discharged in the waste waters.

Chemical separation plants are usually located so that the dilute radioactive wastes can be safely disposed of without incorporating any particular hazard to human food supplies. However, it is customary to make it a condition for discharge of any radioactivity that the biological monitoring of the environment, into which the radioactivity is discharged, is carried out sufficiently extensively and thoroughly both as regards the planning of the sampling and the conduct of the measurements as to be able to demonstrate that no hazard arises to any person from this procedure (13).

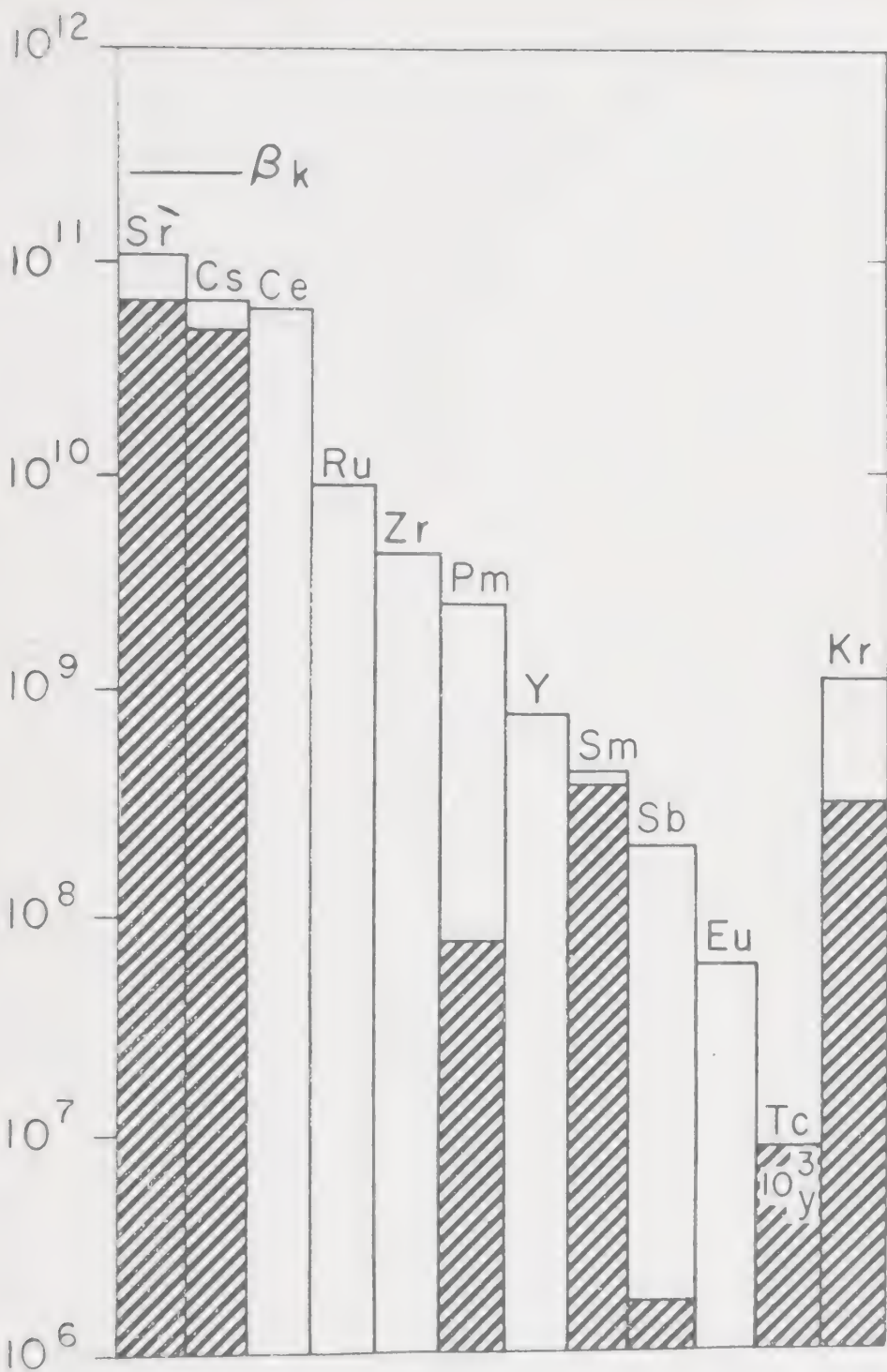


FIGURE 1. Equilibrium activities of individual fission products built up by the annual use of 1,000 tons of fissile materials.

Source: United Nations International Conference, Geneva, Paper P/398, Vol. 9, p. 3-8, by E. Glueckauf, 1955.

KEY



After one year's storage before disposal



After 20 years' storage before disposal

For comparison, total β activity of K^{40} of the oceans

Figure 1 shows the proportions of different nuclides in a storage area which has been operated for a sufficiently long time for the levels of radioactivity to establish an equilibrium with the radioactive decay. The shaded area shows the proportions remaining after 20 years decay. The tabulation is made on the basis of 1,000 tons of fissionable material (e.g., U-235) fissioned per annum in a large world-wide nuclear power system. The chart also shows the amount of radioactive potassium in the ocean purely for comparison. Figure 1 again indicates the biological importance of strontium and cesium isotopes from the viewpoint of any food chain hazards which could remotely arise from long-term storage.

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RADIOACTIVE CONTAMINATION OF SOILS AND CROPS

Index

1. GENERAL INTRODUCTION

2. NATURAL SOURCES OF RADIOACTIVITY

- 2.1 Carbon-14
- 2.2 Potassium-40
- 2.3 The radium, thorium and actinium series
- 2.4 Other natural radioactive elements

3. LEVELS, DISTRIBUTION AND REDISTRIBUTION OF FISSION PRODUCTS

3.1 Strontium-90

- 3.1.1 Regional comparison of Sr-90 deposition
- 3.1.2 Trends in time of Sr-90 deposition
- 3.1.3 Sr-90 deposition and rainfall

3.2 Strontium-89

3.3 Cesium-137

3.4 Redistribution

- 3.4.1 Movement by wind
- 3.4.2 Movement by water
- 3.4.3 Vertical redistribution

4. STRONTIUM-90

- 4.1 Plant physiological aspects
- 4.2 Behavior of Sr-90 in soil
- 4.3 Mechanisms whereby plants are contaminated with Sr-90 under conditions of continuous deposition

- 4.3.1 Qualitative evidence of the extent to which recently deposited world-wide fallout is responsible for the contamination of plants with Sr-90
 - 4.3.2 Quantitative assessments of the extent to which recently deposited world-wide fallout is responsible for the contamination of milk with Sr-90
- 4.4 Assessment of future situations
- 4.5 Effects of management practices
 - 4.5.1 Application of lime
 - 4.5.2 Application of gypsum
 - 4.5.3 Application of fertilizer
 - 4.5.4 Additions of organic matter
 - 4.5.5 Tillage practices
- 5. CESIUM-137
 - 5.1 Plant physiological aspects
 - 5.2 Behavior of Cs-137 in soils
 - 5.3 Entry of Cs-137 into plants
- 6. ISOTOPES OF IODINE
- 7. OTHER RADIOACTIVE SUBSTANCES
 - 7.1 Fission products
 - 7.2 Induced activities
 - 7.3 Plutonium
- 8. REMEDIAL MEASURES FOR RADIOACTIVE CONTAMINATION OF SOILS AND CROPS
 - 8.1 Decontamination of land
 - 8.2 Changes in farming systems
- 9. SELECTED REFERENCES

1. General introduction

The fission products which are of primary concern in food chain studies contrast greatly in their behavior in plants and soils, but it is possible to make some generalizations which are applicable to all radioactive contaminants. When radioactive materials are present in the atmosphere they will be deposited on the earth's surface either by direct deposition or in rain. Thus vegetation and the bare soil will be contaminated. Part of the materials will be deposited on leaves and will subsequently be washed down to the soil surface by successive rains.

The extent to which world-wide fallout is deposited varies with latitude and with rainfall but is not constant throughout the year or during the growing season. In periods of high rainfall the rate of deposition is greater; other meteorological factors also influence this rate.

This irregular supply of radioactive materials causes considerable complications in predicting the levels of contamination of vegetation.

After fission products have been deposited on the earth's surface, they will be redistributed not only between plants and the soil on the same area of land but they will also be displaced to varying extents over the land surface due to the following causes: movement in wind or in run-off, displacement in snow, or through the action of man and animals. In considering the fate of fallout it is therefore important to distinguish the *recent* deposit which will be present at or near the site of initial deposition from older material which has been re-distributed. The form in which fission products are deposited will influence their original distribution. For example, material occluded in rain or released as a vapor will be more readily retained on leaves than large particles such as may be released in accidents. The physical form of fallout will also influence the pattern of re-distribution.

Fission products may enter plants by the following mechanisms:

- (a) *Foliar contamination.* Part of the fission products deposited on the leaves of plants will adhere to their surfaces, part will be absorbed if the deposit is soluble. Both categories will contribute to the contamination of the diet of man (e.g., leaf vegetables) or cattle, (e.g., grasses).

- (b) *Floral contamination.* The inflorescences of plants may be contaminated in the same manner. This will be particularly important when the structure of the inflorescence is such that the current deposit is readily entrapped. If contamination occurs shortly before grain crops reach maturity, absorption will occur mainly in the periphery and in consequence the amount of fission products in diet will depend largely on the extent to which the outer layers of the grain are removed in milling.
- (c) *Plant-base absorption.* Part of the material may reach the basal parts of the plants or roots so near the soil surface, that it is absorbed relatively undiluted with elements from the soil; entry by this route is defined as "plant-base absorption". No precise definition of the sites of absorption can be made since, depending on the character of the plant, the relative extents to which absorption occurs through leaf bases, basal parts of stems, or surface roots may vary greatly. The quantity of material which lodges in this reservoir will vary greatly depending on the nature of plants, cropping systems and on regional conditions. In annual species, the reservoir will contain only material which has been deposited since the crop was planted. In many cases, little will be intercepted in its downward passage to the soil. In perennial crops, for example, pastures, considerable amounts of material may, however, be entrapped in this way and retained for lengthy periods. The extent of retention is likely to be greatest when there is a considerable "mat", consisting of the basal parts of stems and surface roots, near the surface of the soil.
- (d) *Absorption from the soil.* Fission products will reach the surface of the soil either directly from the atmosphere or by downward transfer from leaves or the plant-base reservoir. With the passage of time they will be slowly incorporated into the soil. A fraction of these materials will be absorbed by roots and transported to other organs such as leaves and seeds.

The contamination of crops may, therefore, be caused by direct contamination (foliar and floral contamination), by indirect contamination (absorption from soil), or by a combination of direct and indirect contamination (plant-base absorption). Recent deposit contributes predominantly to foliar and floral contamination whereas the

extent of the old deposit will predominantly govern absorption from material incorporated in the soil. The importance of this distinction is that only the old deposit will have equilibrated with the soil system. There is evidence (*see* sections 4-6 below) that at the present time recently deposited world-wide fallout is an important source of the contamination of vegetation. In the future, however, absorption from the older material accumulated in the soil should become proportionately more important on account of its increasing magnitude relative to the recent deposit.

Although no direct information is available, it appears possible that in dry climates where little rain occurs during the growing season, a considerable re-suspension of material from the soil surface may contaminate the plant shoots; entry may occur in dew.

The factors which control absorption from the soil are numerous and complex. The manner in which radioactive nuclides are distributed in the soil profile is of considerable importance. In undisturbed areas the highest concentration is found at the surface of the soil for a considerable period after deposition; the concentration decreases with increasing depth. The extent of penetration is determined by the texture and structure of the soil, by the concentration of electrolytes in the soil solution, and by the rate at which water moves from the surface to greater depths. Rapid penetration is favored by a low content of clay and humus, a high concentration of electrolytes in the soil solution and by rapid movement of water (11).

Other factors being equal, the extent to which radioactive contaminants are absorbed by plant roots will, therefore, be largely influenced by the rooting pattern. When, however, land has been recently disturbed by deep cultivation such differences may be considerably reduced.

Apart from the concentration of radioactive materials in the different soil horizons, the chemical form of these elements is of great importance. Although recently deposited world-wide fallout dissolves rapidly in the soil solution, its availability to plants may be subsequently reduced in consequence of a number of physical chemical processes. The extent of such reactions differs markedly between different nuclides, for example Cs-137 is rapidly rendered unavailable to plants in many soils; this does not occur with Sr-89 and Sr-90.

By far the most important processes in soils which generally affect

absorption are, however, the exchange reactions which control the distribution of ions between the soil solution and the solid phase. In this regard, the clay and humus content of the soil, the pH , the total concentration of salts and the moisture content of the soil are of considerable importance. In assessing these relationships, it is not necessary to take into account the concentration of the added fission products as it will not be sufficient to alter the existing equilibria.

When fission products are deposited as insoluble particles their incorporation into the soil complex will be considerably slower. A slow release of material into solution will usually occur.

When the equilibrium conditions of a particular soil are known, it is possible to predict the behavior of ions in it. This equilibrium may depend mainly on the concentration of a single ion. For example, because of the similarity of strontium and calcium, the behavior of strontium will be governed by the factors which control that of calcium. In consequence, the absorption of strontium by plants will be determined by the ratio of strontium to calcium in the soil solution.

The principles which govern the absorption of nutrient elements from fertilizers also control the absorption of fission products from the soil. It is, however, difficult to predict the exact quantity of any nutrient which will be absorbed each year because of fluctuating climatological conditions. The same problem arises when an attempt is made to predict the amount of fission products which will be absorbed by agricultural crops in the near future: indeed the problem is still more complex because of the variable pattern of deposition.

When statements in this report are not supported by references, the sources can be found in the working papers of the Committee.

2. Natural sources of radioactivity

Many radioactive elements occur naturally in the earth's crust and in the atmosphere. They enter into the food chain to varying extents. If the consequences of the contamination of the environment with artificial radioactivity are to be adequately assessed, it is necessary to take account of these natural sources.

2.1 CARBON-14

C-14 is formed by the bombardment of nitrogen in the atmosphere with cosmic rays. This naturally formed C-14 is in equilibrium with stable carbon except insofar as recent releases of carbon from fossil fuels have affected this relationship. C-14 amounts to approximately 10^{-10} to 10^{-11} percent by weight of C-12. The production of C-14 in nuclear weapons trials is referred to in a later section.

2.2 POTASSIUM-40

The upper layers of the earth's crust contain approximately 2.5 percent of potassium of which 0.026 percent is the radioactive isotope K-40. The quantity of potassium in agricultural soils varies greatly depending both on the origin of the soils and on the addition of fertilizers containing potassium. It has been calculated that when soils contain 1 percent of potassium, a common level in many localities, the content of K-40 in the upper 30 centimeters of soil is approximately 8 curies per square kilometer. Up to 2 percent of the dry matter in vegetation consists of potassium. Thus, for example, alfalfa hay, an important source of animal forage in North America, may contain approximately 2.5 $\mu\mu\text{C}$ K-40 per kilogram dry weight. The potassium content of vegetables eaten by man is highly variable, but the general level of K-40 is indicated by the fact that one kilogram of fresh cabbage may contain 0.5 $\mu\mu\text{C}$.

2.3 THE RADIUM, THORIUM, AND ACTINIUM SERIES

The decay patterns of these three series of heavy elements are extremely complex. In each case a number of radioactive substances of highly varying half-lives are formed as a result of the emission of alpha and beta particles and gamma rays. All these substances are slowly absorbed by plants; few quantitative data are available. The greatest attention has been paid to isotopes of radium, for example Ra-226, because this element appears to be more readily absorbed than the majority of the other members of these series. It has been estimated that the quantity of radium in soils in North America and in Germany in the rooting zone may range from 15 to 300 millicuries per square kilometer. Considerably higher levels may occur in localized areas.

2.4 OTHER NATURAL RADIOACTIVE ELEMENTS

Radioactive isotopes of the following elements occur naturally: rubidium, lanthanum, samarium, and ruthenium. Very little information is available regarding the absorption of these elements in plants, though it is known that they are present in considerably lower concentrations than K-40, the most abundant naturally occurring radioactive isotope.

3. Levels, distribution and redistribution of fission products in soils

3.1 STRONTIUM-90

Three different methods have been employed for the determination of the accumulative total and the rate of deposition of Sr-90:

- (a) soil analysis;
- (b) gummed film techniques;
- (c) the collection of total precipitation (pot collection).

- (a) Soil analysis is most suitable for the determination of the cumulative deposit of Sr-90 and of the rate of deposition over relatively long periods of time. One of the main problems of this method is locating undisturbed sampling sites and in avoiding sampling at locations where redistribution has occurred.

The most extensive soil data on a worldwide basis have been reported by Alexander (1), who has estimated the total quantity of Sr-90 which reached the ground as "fallout" since the first nuclear explosion. Measurements have been made in many other countries also.

- (b) The gummed film technique produces information on the total radioactivity accumulated from air. It is, however, of limited value for determining the cumulative total of Sr-90, or the rate of deposition, because no radiochemical determination of Sr-90 is made. This can, however, be estimated from the total radioactivity if the age of the deposit is known. No use has been made of gummed film data in this report.

- (c) For the determination of the rate of deposition of Sr-90 over relatively short periods of time, the collection of precipitation has proved to be a very suitable technique. In order to arrive at an estimate of the cumulative deposit, the Sr-90 values of the daily, weekly, or monthly collections are added together. This procedure has the disadvantage that the sampling and analytical errors involved in the collections tend to accumulate in the figure for the cumulative total. For this reason, these data may give a less reliable estimate of the total deposition of Sr-90 than soil assay. This work is now carried out on an extensive scale; the majority of the precipitation collections were started in the course of 1957/58.

3.1.1 *Regional comparison of Sr-90 deposition*

The soil data have been arranged according to 10° latitude bands (1). Figure 2 shows the distribution of Sr-90 in 1958. The major part of Sr-90 deposition occurred in the northern hemisphere, with a maximum between 20 and 60°N and a minimum in the equatorial zone. This distribution is very similar to that in 1956; it is supported by precipitation measurements (13).

The 1958 Sr-90 soil data for some United States of America sites showed levels that are frequently above the world average for the corresponding latitude band.

The average difference between values for the Sr-90 content of soil at sites between one and forty kilometers apart, is of the order of 0.4 millicurie/square kilometer in soils with an average value of 5.2 millicurie/square kilometer.

It should be borne in mind that all the cited data refer to Sr-90 levels at undisturbed sites. The redistribution of Sr-90 due to the topographic characteristics of an area or to variation in deposition due to differences in climatological conditions may result in considerably lower or higher Sr-90 levels than those reported. For agricultural purposes it is necessary to determine the amount of Sr-90 in cultivated soils.

3.1.2 *Trends in time of Sr-90 deposition*

The annual rate of deposition has been reasonably constant from 1954 through 1957. The average yearly increase at Milford Haven (United Kingdom 51.4°N) which is approximately 2.5 milli-

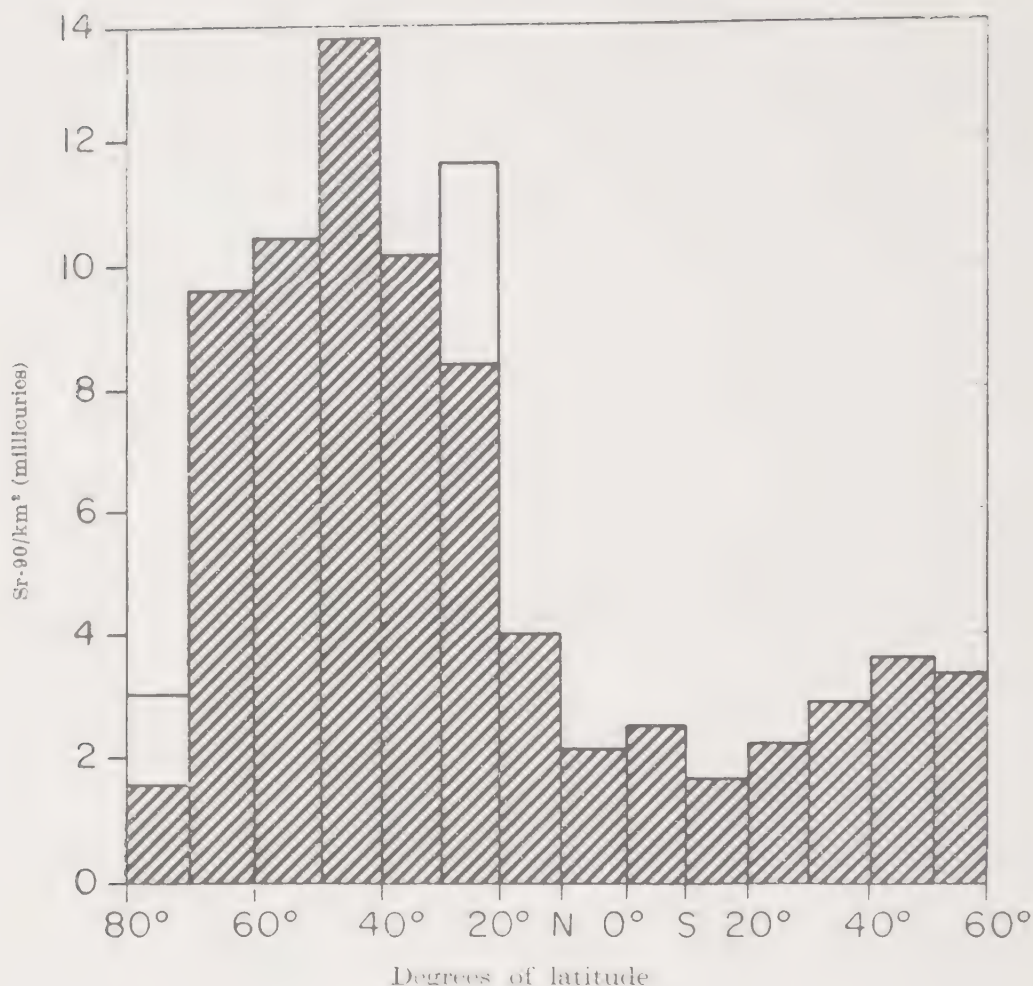


FIGURE 2. - *Millicuries Sr-90/square kilometer in soil - 1958. Source: Alecanler. The nonhatched bars in the figure represent anomalous values. See text (1).*

curies Sr-90/square kilometer, is about the same as that suggested by the soil data at the corresponding latitude (1). In New York City (41.6°N) the average yearly Sr-90 increase is higher, about 4 millicuries/square kilometer. During 1958, an increase in the annual rate of deposition was observed from both United States of America and United Kingdom data, presumably due to the intensified nuclear device testing of the United States of America and the Union of Soviet Socialist Republics in the latter portion of that year.

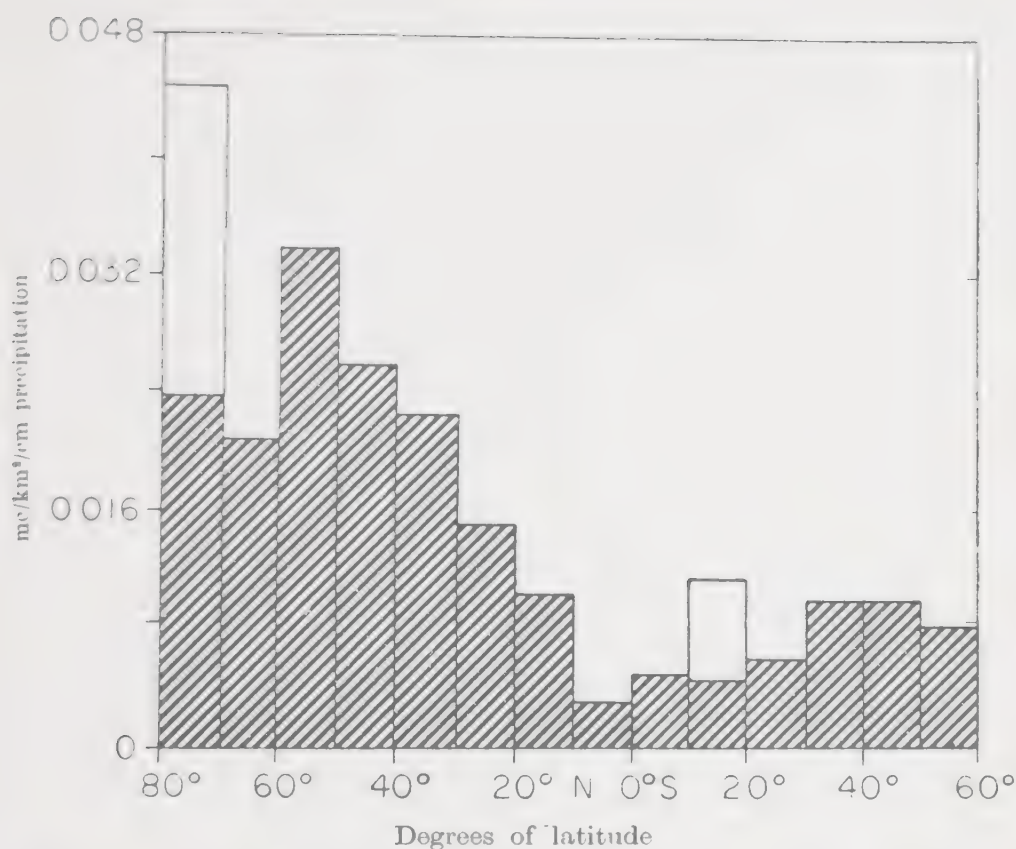


FIGURE 3. - Rate of Sr-90 deposition mc/km²/cm. of precipitation, 1953-1958. Sources: Alexander. The nonhatched bars in the figure represent anomalous values. See text (1).

The monthly measurements of Sr-90 in precipitation at New York City and Milford Haven show fluctuating values (13; 16). In a few cases, weekly strontium fallout data have been published (New York and Pittsburgh, United States of America); these indicate that in many instances the deposit in one or two weeks contributes the majority of the monthly total.

3.1.3 Sr-90 deposition and rainfall

Various attempts have been made to relate Sr-90 deposition to precipitation. Figure 3 shows that the rate of deposition in mc/square kilometer/centimeter of precipitation in 1956-58 is not equal for all

latitude bands. Particularly in the equatorial region, the Sr-90 content of the rain is very low as compared with the northern hemisphere. Extensive studies of the relationship between the monthly Sr-90 deposition and the amount of precipitation for various sites have been made (13, 4). Coefficients of correlation between Sr-90 deposition and rainfall for various United States of America sites show that the degree of correlation fluctuates with the month of sampling and latitude of the sites. Within limited areas the total precipitation is more systematically related to the Sr-90 fallout than it is throughout the whole ten degree latitude belt. Within belts, the drier sites have a higher deposition of Sr-90 per unit of rain than do the wetter ones.

3.2 STRONTIUM-89

The accumulation of Sr-89 is of little importance because of its short half-life (53 days). The rate at which it is deposited is mainly dependent on the rate at which the troposphere is contaminated. However, Sr-89/Sr-90 ratios in soil and agricultural materials are useful characteristics for establishing relative dates of contamination with Sr-90.

3.3 CESIUM-137

As opposed to Sr-90, few data have been reported on the levels of Cs-137 in soil. Because of the proportionately constant fission yields of Sr-90 and Cs-137, an estimate of Cs-137 levels in various latitude bands may, however, be obtained by multiplying the Sr-90 soil data by the factor 1.6 – 1.8 (13, 5).

From the standpoint of a material balance of the total amount of Sr-90 that has been deposited on the earth, the greatest uncertainty is due to the fact that little information is available regarding the deposit on the oceans, which occupy more than two thirds of the earth's surface. Samples from oceanic island sites, where rainfall is not influenced by elevation, indicate, however, that the fallout of Sr-90 is similar per centimeter of rainfall to that on the land masses in the same latitude.

3.4 REDISTRIBUTION

3.4.1 *Movement by wind*

In the very cold, very dry northern areas vegetation is sparse and in small clumps only. The soil materials blow and shift about with the winds. In the winter snow accumulates in the clumps of vegetation and also in distant drifts. Abnormal accumulations of Sr-90 thus occur in the clumps. Perhaps there is no satisfactory way of estimating the cumulative deposition in these partially vegetated, wind-swept areas.

A similar situation exists in semi-desert and bunch grass areas. Between the clumps of plants the soil surface horizon is in frequent motion due to wind-action. Blown materials accumulate in the vegetation at elevations greater than those of bare areas. Infrequent heavy rains wash some of the soil material in the clumps back into the lower-lying bare spots. Several fold differences have been found between replicate samples taken in such areas at some distance from the Nevada Test Site. This kind of local variation is to be expected where the vegetative cover is not complete and the movement of soil material by wind action is common (2).

In the Temperate Plains areas a comparable situation exists on a much larger scale. Snow blows away from the unsheltered open areas and accumulates in the fence rows, gulleys, and canyons. It is virtually impossible to find areas that have on them all the Sr-90 that fell there and none other.

3.4.2 *Movement by water*

Water movement on the land surface and soil erosion contribute to the re-distribution of fallout. How great the nonuniformity resulting from this redistribution is has not been determined but some recent work shows that on soil erosion study plots at La Crosse, Wisconsin and Tifton, Georgia, about 99 percent of the Sr-90 falling on the soil remained on the plot (6). But the material lost by runoff was about ten times as high in Sr-90 per unit of weight as the plow depth soil remaining. This could, of course, result in accumulation of Sr-90 in small areas at the foot of slopes and in alluvial positions of from several times to one order of magnitude higher than those found in the surrounding areas.

The movement of Sr-90 by erosion will not only cause wide variation in the magnitude of the deposit within a region, but it may also cause a lowering in the mean level insofar as Sr-90 passes into streams and rivers.

3.4.3 Vertical redistribution

The vertical distribution of Sr-90 in soils is of concern because of its importance in determining the uptake of Sr-90 by plants. In the main Sr-90 has remained close to the surface of vegetated undisturbed soils.

Table 4 shows results for two very sandy and porous soils in which downward movement due to leaching would be expected to be greatest; even here movement was slow.

TABLE 4. - VERTICAL DISTRIBUTION OF Sr-90 IN TWO SANDY SOILS

Depth, in cm.	Plainfield sand, Illinois, millicuries per square km.		Bayboro loamy sand, Georgia, millicuries per square km.	
	1957	1958	1957	1958
0-2 0-5	9.1	10.5	6.7	8.1
2-6 5-15	3.0	5.8	0.9	3.1
6-12 15-30	0.5	2.0	0.3	1.0

There are, however, many cases of unplowed soil where Sr-90 has penetrated to greater depths; for example, in soils having a great turnover due to earthworms, termites and other organisms and in soils that form cracks on drying. Cultivation, of course, results in mixing the Sr-90 to the depth of cultivation. The degree of homogeneity obtained depends on the nature of the cultivation.

Compost and sewage sludge are sources of Sr-90 that may be more concentrated than the soil on which they are used. Heavy use of sewage sludge in particular may result in a substantial increase of this radionuclide on farmland. It is, however, used only on limited areas.

4. STRONTIUM-90

In considering the entry of Sr-90 into food chains, the ultimate objective is to estimate the internal dose of radiation which will be received by the skeleton of man or animals. This will depend primarily on the ratio of Sr-90 to Ca in the total diet. Thus, the examination of the quantities of Sr-90 and calcium in agricultural products is of particular importance. It is convenient to express results in terms of the "Strontium-Calcium Observed Ratio (OR)", defined thus:

$$\text{OR}_{\text{sample/precursor}} = \frac{\text{Sr-90/Ca sample}}{\text{Sr-90/Ca precursor}}$$

If measurements of the observed ratio are to be of value it is essential that the ratio measured in the precursor truly reflects the relative availability of the two ions for absorption into the biological system under study. This subject is discussed in greater detail in Appendix 3.

4.1 PLANT PHYSIOLOGICAL ASPECTS

When plants are grown in soil the quantities of strontium and calcium they absorb reflect the behavior of the two ions both before and after they enter the plants. It is, therefore, convenient to consider first the results which are obtained when plants are grown in simple inorganic solutions. Many such studies have shown that Sr-90 and calcium are absorbed in a similar manner, thus the OR plant over medium is close to 1. Calcium thus effectively "dilutes" strontium in the absorption process; and thus the rate at which strontium is absorbed is inversely related to the concentration of calcium in the rooting medium.

Detailed plant physiological studies show that the rates of movement of strontium and calcium within plants are not identical; the magnitude of such differences, however, is usually not sufficiently great to be of importance in food chain studies.

On theoretical grounds it might be expected that the movement of Sr-90 would be more closely related to that of stable strontium.

In practice, however, the quantity of calcium in plants exceeds that of stable strontium by so large a factor that the absorption of both the radioactive and stable isotopes is effectively controlled by the level of calcium.

The movement of strontium through plants is predominantly unidirectional from the roots upwards. Thus, Sr-90 which has entered leaves is little redistributed to other tissues. This is of considerable importance in assessing the consequences of the aerial contamination of vegetation: Sr-90 directly absorbed by leaves which are not consumed by man or animals is of little importance from the viewpoint of food chain studies.

4.2 BEHAVIOR OF SR-90 IN SOIL

The slow penetration of Sr-90 into the soil causes the mean ratio of Sr-90 to calcium to which the roots of plants are exposed to vary depending on their rooting depth. Shallow-rooted crops thus show the highest ratios of Sr-90 to calcium assuming that the distribution of calcium is uniform throughout the soil.

A further complication in elucidating relationships between strontium and calcium in soil comes from the fact that conventional procedures for estimating "available" or "exchangeable" calcium may not give an adequate measure of the quantity of soil calcium with which the strontium is effectively diluted on absorption. Evidence from numerous investigations, however, indicates that the two ions are absorbed in proportion to their true availability in the soil. Thus, if the ratio of Sr-90 to calcium in the equilibrium soil solution is estimated, the ratio in which the two ions will be absorbed can be inferred.

4.3 MECHANISMS WHEREBY PLANTS ARE CONTAMINATED WITH SR-90 UNDER CONDITIONS OF CONTINUOUS DEPOSITION

Four mechanisms of contamination have been defined: —

- (a) foliar contamination;
- (b) floral contamination;
- (c) plant-base absorption;
- (d) absorption from the soil.

The first two mechanisms can result in contamination with Sr-90 which is either deposited on surfaces or absorbed by the tissues. Both types of contamination can be caused only by Sr-90 deposited during the growth of the leaf or inflorescence in which it is found; they are, therefore, due to material deposited only in the recent past. The same may be true of plant-base absorption with annual plants. With perennial species, however, it is possible that Sr-90 which has been deposited at any time during the growth of the plant may enter by this route. The "apparent age" (an integration of time between deposition and absorption) of Sr-90 which enters by plant-base absorption will depend on the extent to which prostrate stems, inrolled leaf bases and surface roots intercept the deposit.

In contrast to the other three routes of entry, absorption from the soil may lead to the contamination of plants with Sr-90 which has been deposited at any time since fallout commenced, although especially with shallow rooted species the more recent deposit will generally be absorbed to the greatest extent.

The possible magnitude of the contamination directly deposited on vegetation is indicated by the fact that 25 percent or more of a material deposited in a finely divided state, or as a vapor, may lodge on the leaves of perennial pastures. In contrast it has been found in the United Kingdom that 0.2 to 2 percent of Sr-90 which is in soil is absorbed by plants each year (8). Given equal quantities of Sr-90 in the plant-base reservoir and the soils, plant-base absorption can be expected to be considerably greater than absorption from soil.

From the viewpoint of the entry of Sr-90 in the food chains, it is particularly important to consider the relative extents to which the Sr-90 absorbed by these routes will be diluted by calcium within plants. The calcium supply in the soil will not affect the quantity of Sr-90 which enters by floral or foliar contamination, but it will affect the ratio of Sr-90 to calcium in plants if it affects their calcium content; the highest ratios will be found in plants growing in very calcium-deficient soils, all other factors being equal. Since the extent to which Sr-90 is absorbed from soil will depend on the degree to which it has equilibrated with calcium before absorption, the resultant ratio of Sr-90 to calcium in plants will be considerably lower and more closely controlled by the calcium content of the soil. Plant-

base absorption will lead to a situation similar to foliar or floral contamination except insofar as calcium may enter the plant-base reservoir; these effects may be considerably altered when crops have been top dressed with lime.

4.3.1 *Qualitative evidence of the extent to which recently deposited world-wide fallout is responsible for the contamination of plants with Sr-90*

The delay period between the deposition of Sr-90 and its appearance in plants depends on the relative magnitude of foliar, floral, plant-base and soil absorption. Evidence on this question is provided both by surveys of the content of Sr-90 in food and other agricultural products and by experimental studies. Since there is little difference between plant species in the extent to which they discriminate between strontium and calcium, the ratio of Sr-90 from world-wide fallout to calcium in all plants growing in the same locality may be expected to be similar except insofar as it is affected by the depth of their roots if Sr-90 were absorbed solely from soil. Furthermore, milk, on which a considerable body of data is available, would be expected to show ratios of Sr-90 to calcium of one seventh to one tenth of those in vegetable foods owing to the observed ratio between the two ions in their passage from the diet of animals to milk. A comparison of the ratios of Sr-90 to calcium in different articles of diet may, therefore, provide some preliminary information as to the manner in which Sr-90 enters plants. In the United Kingdom during 1958, where samples were collected throughout the country, the ratios of Sr-90/Ca were milk 7, whole grain 51, and leaf vegetables 8.7, $\mu\mu\text{c}/\text{gram Ca}$. These results indicate that the Sr-90 in the produce was largely due to direct contamination.

Some information from other parts of the world is summarized in Appendix V, Table 7 of this report, and suggests that a similar relationship occurs in other regions; however, the fact that the samples were not in all cases collected from the same localities makes this comparison less certain.

More detailed information on the mechanism whereby fission products enter diet can be obtained from the examination of ratios of Sr-89 to Sr-90 observed in such surveys. In any sample of fission

products this ratio must fall by a factor of 2 approximately every seven weeks on account of the shorter half-life of Sr-89.

The ratios of Sr-89 to Sr-90 in milk produced in the United States of America and in Canada during 1957 and 1958, and in the United Kingdom in 1958, were usually in excess of 8 during the summer months, and sometimes attained considerably higher values after weapons tests; during the winter months, low values (1 or 2) were observed (10). In the United Kingdom, it was possible to compare the ratios in milk with those in the current rainfall; in the summer of 1958 the ratio in milk generally exceeded one half of those in rain (15). Thus, the "apparent delay period" in the passage of Sr-90 to milk was one to two months. Measurement of the short-lived barium-140 (half-life 12 days) in the United States of America soon after weapon trials in 1957 and 1958 provide further evidence of the appearance of relatively recently deposited fission products in milk.

The contribution of recent fallout to the contamination of leafy annual vegetables is again shown by the examination of ratios of Sr-89 to Sr-90. Values equivalent to half or more that of current fallout were found for the inner and protected leaves of cabbages in the United Kingdom during 1958 (15). Since these tissues are not exposed to direct foliar contamination, the deposition of Sr-90 in the cups formed by leaf bases, and its entry by plant-base absorption, appear to be reasonable interpretation.

Attention has already been directed to the higher ratios of Sr-90 to calcium in grain than in other plants grown in the same localities. Evidence that this is attributable to floral absorption is forthcoming from three sources. Studies in Japan have shown that the higher ratio of Sr-90 to calcium in grain than in other tissues of rice plants, which are observed from world-wide fallout, does not occur in experiments where radioactive strontium has been added to the soil; direct contamination has been inferred (14). An examination of the ratios of Sr-89/Sr-90 in different tissues of grain from different parts of the world points to the conclusion that variable but frequently large fractions of the total Sr-90 in grain may have entered by direct absorption (15). Laboratory experiments in which Sr-89 was applied as a fine spray to crops at different stages of development have shown that when wheat is sprayed after the ears have emerged, the resultant ratios of Sr-89 to calcium in the mature grain may be 20 or more

times greater than that observed when the spray is applied before ear emergence: the protection of the emerged ears with polythene sheaths during the spraying operation eliminates this effect (7).

The foregoing information does not, however, enable quantitative conclusions to be drawn with regard to the extent to which recently deposited Sr-90 is responsible for the current contamination of the total diet. For example, the ratios of Sr-89 to Sr-90 observed in milk in the United Kingdom are equally compatible with the contrasting interpretations that all the Sr-90 was deposited less than two months previously or that it was equally divided between very recent and very old material from the soil.

Highly variable relationships between the contamination of vegetables and the quantities of Sr-90 deposited in the preceding months must, however, be expected. This is particularly obvious with respect to grain crops. The same quantity of Sr-90 deposited may have very different effects depending on whether it falls before or after the emergence of ears. The extent of rain is another major variable since it has been shown that recently deposited material is readily removed from leaves in rain. Furthermore, all environmental factors which affect the growth of plants may influence the extent to which Sr-90 is retained on the leaves or in the "plant-base" zone. Considerable differences are to be anticipated in different seasons and among different regions.

4.3.2 Quantitative assessments of the extent to which recently deposited world-wide fallout is responsible for the contamination of milk with Sr-90

The discussion of quantitative relationships is here restricted to milk because considerably less information is available on other foods. If the long-term effects of world-wide fallout are to be predicted, it is necessary to estimate the relative extent to which the quantity of Sr-90 in diet in the future will depend on the current rate of fallout as opposed to the cumulative total in the soil. Attempts have been made to relate these quantities by equations of the following type: —

$$C = a.D_p + b.D_t$$

when C is the ratio of Sr-90 to calcium in a biological material, D_p and D_t are the annual rate of fallout and the mean cumulative total

respectively, and a and b are proportionality factors. Since the time at which fallout is deposited, and meteorological factors may greatly affect the extent to which plants are contaminated by recently deposited material, the value of a must be expected to vary considerably from year to year. This, however, does not mean that the method should be dismissed as inapplicable for, if sufficient data were available, it would be possible to estimate its mean value. A further difficulty arises from the fact that the extent to which Sr-90 is absorbed from soil may slowly decrease as a result of its more thorough incorporation; it cannot be assumed that b is constant. The importance of assessing future relationships makes it, however, necessary to consider what information can be obtained despite these limitations.

The longest continuous series of observations of Sr-90 in milk from a single source is for a milk drying plant at Perry, New York. An assessment of the proportionality factors in an equation similar to that quoted above was attempted in 1957 and quoted in the Report of the United Nations Scientific Committee (17, 3). The calculation has now been repeated using the data available up to May 1959.

The two estimates of the proportionality factors are: —

<i>Estimate based on results</i>	<i>a</i>	<i>b</i>
1954-1957	0.23	0.34
1954-1959	0.41	0.24

The confidence intervals for the more recent estimate are: $0 < a < 1.08$ and $0 < b < 0.49$ ($P = 0.05$).

An alternative and theoretically more attractive method for assessing such relationships is by multiple regression analysis in which the weekly or monthly levels of Sr-90 in milk are expressed as functions of the cumulative deposit and of the weekly or monthly deposit. This has been attempted in an analysis of the results of the New York City milk examined by the Health and Safety Laboratory of the United States Atomic Energy Commission. This investigation which will be described in detail elsewhere leads to the conclusion that the present data are inadequate for this purpose (3). When

however, considerably more data are available, it may be possible to obtain information by both of these methods.

An alternative approach for assessing the relative importance of recent fallout and the cumulative total in determining the content of Sr-90 in milk has been attempted in Germany. The increments of Sr-90 in arable land have been compared with those in pastures during 1959 and it has been concluded that some 20 to 30 percent of the Sr-90 reaching milk had been absorbed from the soil. This is a preliminary approximation only (12).

In the United Kingdom, field-scale experiments have been undertaken to estimate the extent to which Sr-90 will be absorbed from soil. The method employed has been to spray Sr-89 onto land which is then cultivated to various depths before crops are sown. The results for experiments carried out in six contrasting areas are now available for two years. They have been used to estimate the average ratio of Sr-90 to calcium in pasture grasses throughout the country which would be expected from the presence of one millicurie per square meter distributed in soil in a manner comparable to the present deposit of world-wide fallout. The estimated value is $1.6 \mu\text{c Sr-90/gram Ca}$ (10). On this basis the level of Sr-90 to be expected in milk in 1958 as a result of absorption from soil has been computed and compared with the levels actually observed during that year, when information was obtained on the content of over 40 percent of the total production in the country. It was concluded that absorption from the soil was responsible on the average for approximately 20 percent of the Sr-90 in milk. It was further estimated that with the passage of time, when the present cumulative deposit of worldwide fallout will be more thoroughly incorporated in the soil, it will lead to 12 percent (10) of the present level in milk.

The tentative nature of this assessment has been stressed by its authors; the manner in which radioactive strontium was applied to the soil could not exactly simulate the distribution of world-wide fallout and data for experiments on a limited number of sites only were available. Furthermore, contrasting relationships are to be expected in other countries and in different years, due both to variations in climatic conditions and the rate of fallout. However, the conclusion reached is compatible with information drawn from the ratios of Sr-89 to Sr-90 in milk during the same period.

4.4 ASSESSMENT OF FUTURE SITUATIONS

Quantitative predictions applicable to diverse areas cannot be made both because of the complexity of the factors which control the contamination of plants and also because the future amount and distribution of world-wide fallout is unknown. Certain generalizations can, however, be made. It is evident that, even if the deposition of world-wide fallout were to continue at its present rate, the contamination of diet would probably increase, but not in proportion to the increase in cumulative total; furthermore, if the rate of deposition declines markedly, an appreciable reduction in the contamination of diet could occur even though the cumulative total in the soil is still increasing. Under either set of circumstances, however, the fraction of the Sr-90 in diet which has been absorbed from the soil will increase. This will bring about changes in the relative extent to which different types of produce are contaminated. The contamination of materials which at present contain the largest proportions of recently deposited materials, e.g., milk and grain in some areas, will fall relative to other articles of diet. Eventually, if the effect of recently deposited material becomes negligible and the deposit in the soil is relatively uniformly incorporated throughout the rooting zone of plants, the ratios of Sr-90 to calcium in all plant tissues will be similar in any one area and the values in milk will be one seventh to one tenth of those in vegetable foods.

So long as the recent deposit makes a considerable although variable contribution to the contamination of diet, it is evident that knowledge of the cumulative total in the soil will provide little information on the quantities of Sr-90 in human diet. This can only be determined by analyzing representative samples of the major foods either individually or combined in the proportions in which they are consumed.

Apart from enabling the present situation to be assessed, surveys of world-wide fallout are a potential source of information for predicting the situations likely to be caused by instantaneous accidental releases.

If the results are to be used for this purpose, it is important to relate the contamination of diet to the quantity of fallout which has been recently deposited.

4.5 EFFECTS OF MANAGEMENT PRACTICES

4.5.1 *Application of lime*

Many investigations have been made of the effects of liming on the uptake of Sr-90 by crops (9). It is currently generally accepted that addition of lime to a particular soil in excess of saturation of the cation exchange complex with calcium is of no benefit for this purpose. Moreover, in order to maintain balanced soil fertility, a part of the exchange complex must be occupied by other nutrient cations. It is, therefore, recommended that acid soils be limed only to the level which will provide optimal productivity. The application of lime to neutral or alkaline soils generally is ineffective.

As an increase in exchangeable calcium is only one of a number of effects of liming of acid soils, a simple relationship between lime status and the radiostrontium content of the crops is not always observed. When a soil is deficient in calcium, lime may affect both the size of the plants and the uptake of radiostrontium. Raising the exchangeable calcium level above that at which this deficiency is remedied increases further the dilution of soil strontium with calcium. It has been concluded from the results of field experiments, however, that the addition of lime to soil will have a large effect only in soils of low calcium status (8).

The result of this dilution effect of liming often is a reduction in the ratio of radiostrontium to calcium in the plant, and not an absolute decrease in the radiostrontium content. Since this ratio appears to be the significant expression of the radiostrontium level in the food chain, however, this particular effect is of value. A reduction in this ratio of about 3 is the maximal effect to be expected, and even so great an effect is not usual. It has also been found that lime applied to the surface of an established grass pasture, at the rate of two tons per acre, can reduce the ratio of radiostrontium to calcium in the grass by as much as about two-thirds. Regardless of the sources of Sr-90 to the plant, the general benefit of liming, where it is effective, is an increase in the calcium concentration in the plant.

4.5.2 *Application of gypsum*

Experimental evidence indicates that additions of gypsum might be of some benefit on saline and alkali soils which have a high degree of saturation with sodium. Replacement of exchangeable sodium

by the calcium of gypsum is a common reclamation treatment for such soils, and this increase in exchangeable calcium should decrease the uptake of radiostrontium or the ratio of radiostrontium to calcium in the plant by virtue of the dilution effect.

4.5.3 *Application of fertilizer*

Fertilizers applied within the normal agricultural range of applications are not expected to have large effects on the uptake of radioactive contaminants. As with liming, the general recommendation is for applications which will provide optimal soil productivity. Fertilization treatments which might increase or decrease the uptake of Sr-90 would have comparable effects on the uptake of calcium. Consequently, the ratio of Sr-90 to calcium in plants probably is not altered greatly by fertilizer practices.

4.5.4 *Additions of organic matter*

Plants have been reported to have lower concentrations of radiostrontium and calcium under experimental conditions when large amounts of organic matter were first mixed in the soil and allowed to decompose partly before planting (9). However, the quantities of organic matter required to produce a substantial reduction are relatively huge, and the effect probably would not remain after decomposition was completed. In this treatment also, only slight changes in the ratio of radiostrontium to calcium in the plants should be expected.

4.5.5 *Tillage practices*

Plowing under of a surface deposit of radiostrontium to a depth of 30 centimeters has been found to reduce by two-thirds the ratio of radiostrontium to calcium in the leaves of ryegrass, while cultivation to 10 and 17 centimeters produced intermediate reductions (8). The effect of depth of tillage on cereal and root crops, however, was of much less importance. It can be concluded that deep cultivation will reduce the radiostrontium content of plants if they are shallow rooted. A possibly effective combination of practices for calcium-deficient grassland might be a lime application on the

grass followed by deep plowing, and possibly followed by tilling of a second lime application into the surface soil.

The ratio of Sr-90 to calcium in paddy rice has been reduced by about one third when a surface application of Sr-90 was mixed into the soil under experimental conditions in Japan (15).

5. Cesium 137

Because of the long half-life of Cs-137 and the fact that it is produced by nuclear fission in a constant ratio to Sr-90, it is convenient to consider its entry into food chains in comparison with that nuclide. Points of contrast will alone be noted.

5.1 PLANT PHYSIOLOGICAL ASPECTS

In contrast with Sr-90, cesium is readily mobile within plants. Cesium which enters leaves is transferred to roots and storage organs and with the passage of time the ratio of cesium to potassium may be expected to become relatively constant throughout the plant.

5.2 BEHAVIOR OF CS-137 IN SOILS

Although cesium belongs to the same group in the periodic table as potassium, the behavior of the two elements in soils contrasts. Cesium is fixed in forms largely unavailable to plants as a result of entrapment in the lattices of certain clay minerals. Its availability to plants is, therefore, low -- one to two orders of magnitude less than that of Sr-90. If, however, soils contain abnormal quantities of natural cesium, the availability of carrier-free Cs-137 could be increased. It has been reported that the addition of fertilizers containing ammonium may decrease the fixation of cesium in some soils; the addition of potassium may also have some effect although it is not usually large.

5.3 ENTRY OF CS-137 INTO PLANTS

Cs-137 may enter plants by direct contamination, or by plant base absorption, as has been described with respect to Sr-90. The contribution from the soil, however, will be considerably lower and

thus the levels in plants may be expected to depend solely on the quantities of Cs-137 deposited from the atmosphere in the recent past. It is of interest that surveys of the contamination of milk with Sr-90 and Cs-137 in Europe and in North America indicate that the content of cesium in milk is usually between 5 and 11 times that of Sr-90. This relatively narrow range may indicate a general similarity in the mechanism of absorption, but considerably fuller information is necessary before a precise conclusion can be reached.

6. Isotopes of iodine

Several isotopes of iodine are formed by nuclear fission: I-131, half-life 8 days; I-132, half-life 2.2 hours; and I-133, half-life 21 hours. If fresh fission products are deposited, the short-lived nuclides will initially make a major contribution to the exposure because of their greater abundance. The effects of I-132 may be greater than its half-life suggests since it is a decay product of Te-132 (half-life 28 hours). When, however, older fission products, or equilibrium mixtures, such as are likely to be present in reactors, are released, I-131 will be the major source of concern.

Because of the short half-lives of the isotopes of iodine, absorption from the soil is unimportant. It will usually be necessary to consider only the foliar contamination of plants with I-131; floral absorption will be of little importance from the viewpoint of food chain hazards in view of the delay which normally occurs between the harvesting of grain and its consumption. Much larger quantities of I-131 may be transferred to human diet through milk than in other ways. Thus, from the practical viewpoint, the question of greatest importance is the extent to which I-131 is retained on the edible tissues of plants which grazing cattle consume. Twenty-five percent or more of the material initially deposited may lodge on herbage of types edible by cattle in European pastures. I-131 may be removed from herbage by rain. After the Windscale accident in the United Kingdom, rain caused the activity of I-131 in grass to decrease in the following two to three weeks, by about twice the extent attributable to the decay of radioactivity. Because of the contrasting ways in which pastures are managed in different countries and the different nature of the swards, it is impossible, however, to make any generalization which is broadly applicable.

7. Other radioactive substances

7.1 FISSION PRODUCTS

If fission products are released in fission yield, isotopes other than those of strontium, iodine and cesium will be minor sources of exposure because of their short half-lives or their slow passage into food chains. The short-lived isotope Ba-140 (half-life 12 days) will be transferred to milk in appreciable quantities when fresh fission products are deposited. Over longer periods it is, however, of minor significance.

A number of the longer-lived fission products will accumulate in the soil to an appreciable extent. For example, four years after fission, Ce-144 and Pr-144 contribute nearly 30 percent of the total radioactivity and the quantity of promethium is similar. At earlier times, Zr-95 + Nb-95 will make an appreciable contribution. These elements are, however, not accumulated to an appreciable extent in biological systems.

7.2 INDUCED ACTIVITIES

C-14 produced by atomic weapons trials has caused the specific activity of atmospheric carbon in the troposphere to be somewhat increased. Thus, C-14 leads only to a very small increase in the natural activity of the food.

A wide range of other induced activities have been released into the environment as a result of the irradiation of materials surrounding atomic weapons or through the release of effluent from reactors. These include isotopes of the following elements; Fe, Na, Mn, Mg, P, Co, and Zn. None of these substances enters food chains to an extent approaching that of the principal source of contamination already discussed.

7.3 PLUTONIUM

It is widely accepted that the primary hazard from plutonium, should it be released into the atmosphere, is likely to be due to inhalation by man or animals. Nonetheless, because of its very long half-life, consideration should be given to its fate when it is deposited on the surface of the land. Such information as is at present available

indicates that when plutonium is introduced into soil it is largely rendered into forms unavailable to plants by "fixation" mechanisms. Furthermore, the rate at which plutonium is absorbed from freely soluble sources is extremely slow and the major part of the plutonium which enters plant roots is retained there, with little reaching the above-ground tissues.

8. Remedial measures for radioactive contamination of soils and crops

Remedial measures for the reduction of radioactive contamination of the food chain should be considered only in emergencies declared by responsible authorities. Decisions concerning the application of remedial measures probably never will be made easily, as they must be based on an attempted weighing of medical assessment of probable injury from radiation against such factors as cost, reduction of food supplies, and the disruption of economic and social patterns. The measures mentioned here include some which might be purposely applied to soils and crops in order to reduce the contamination of land and land products.

8.1 DECONTAMINATION OF LAND

Methods of this nature are recommended only for cases of acute contamination of land by Sr-90. Those which have been investigated include intensive leaching with neutral salts, acid salts, and acids; removal of contaminated sod, crops, and crop residues; scraping of several inches of contaminated surface soil; and applying asphalt and removing the hardened crust from the soil surface (9). Soil structure and fertility obviously would be damaged by some of these measures.

8.2 CHANGES IN FARMING SYSTEMS

Change in crop and animal production systems, as also for land decontamination, are recommended only for cases of acute contamination of land by Sr-90. The selection of alternative farming systems therefore would depend on the extent to which the content of soil derived Sr-90 in the diet of the population might be reduced, particularly that of the younger people.

Additional factors would have to be considered in deciding the feasibility of particular changes involving a considerable fraction of a community's farmland. Agricultural aspects include the adaptation of specific crops to specific soils, climatic factors that influence crop and animal production, and the ravages of insects and plant disease. Disruption of marketing systems, transportation costs, and need for processing facilities represent some of the economic considerations. Personal food consumption preferences comprise a further obstacle. It is recognized that in many areas the possibilities of land-use changes would be extremely limited.

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MOVEMENT AND BEHAVIOR OF RADIOACTIVE MATERIALS IN ANIMALS AND ANIMAL PRODUCTS

Radionuclides enter the human body primarily via the food chain, of which animal products are an important link.

The objectives of this report are to consider present knowledge of the movement of certain nuclides into foods of animal origin. This would include knowledge of the sources of these nuclides to animals, their metabolism and concentrations in the tissues and fluids.

Special attention is given to the relations that exist between the quantity of a radionuclide in the animal body, the content in specific tissues and fluids and the quantity of nuclides which enter the animal.

This report is restricted primarily to the consideration of Sr-90, Cs-137 and I-131, but will include such information as is available pertinent to domestic animals on other radionuclides that may be of importance.

The effect of exposure to radionuclides on the well-being of the animal is not considered in this report because it will be of minor practical importance as compared to that of man in spite of the fact that grazing animals will acquire higher body burdens than will human populations. A study of exposure effects, if feasible, on domestic animals may lead to a better understanding and anticipation of effects on the human population.

Management and environmental variables

There are many factors that could influence the degree of exposure of farm animals to radionuclides. The most important of these include:

- (a) types of pasture;
- (b) extent of barn feeding;

- (c) miscellaneous feeding practices such as age of food, degree of supplemental feeding and use of barn-stored food;
- (d) source of water;
- (e) housing practices, e.g., degree of sheltering of animals from surface contaminants.

Because of the variables governing the degree of contamination of animals it can be readily understood why one may expect large variation in degree of contamination of animal food products, even from apparently similar sources.

Strontium

Long-term effects of radioactive strontium are related to Sr-90. From the standpoint of studies involving mechanism of short-term effects very often Sr-89, Sr-85 or stable strontium are measured or employed. The following considerations of mechanism would apply to any isotope of strontium properly used.

The concentration of Sr-90 in a tissue, secretion or food, is the essential information for evaluation of Sr-90 hazard. It appears that consideration of strontium-calcium ratios is of much greater usefulness than consideration of strontium retention. The reasons can be summarized:

- (a) There appears to be a reasonably constant relationship under normal dietary conditions between strontium-calcium ratios in diet and those in the body or milk.
- (b) The strontium-calcium ratio predicted in the body at any given time will reflect the radioactive contamination at the local areas in bone that are metabolically active at the time.
- (c) Radiostrontium retentions will vary greatly with age, diet, and other factors: in mature individuals retention will progressively decrease to zero as the skeleton equilibrate with Sr-90 under constant intake; calcium retentions would be expected to be parallel.
- (d) The metabolism of radiostrontium ingested will be governed by the calcium intake.

- (e) The calcium levels of the body and tissues are reasonably constant regardless of dietary intake under adequate or optimal nutritional conditions and the calcium of blood is under homeostatic control. This behavior of calcium makes it a good standard of reference for expression of strontium levels.
- (f) Calcium metabolism and nutrition have been widely studied in the past.

Investigations of strontium metabolism have shown that, in the movement of a mixture of strontium and calcium across physiological barriers, calcium moves at a relatively different rate from that of strontium. This differential movement for given barriers appears reasonably constant.

The term used to express this differential behavior of strontium and calcium is the "Sr-Ca Observed Ratio" or OR and is defined as follows:

$$\text{OR}_{\text{sample/precursor}}^1 = \frac{\frac{\text{Sr}}{\text{Sr} + \text{Ca}} \text{ of sample}}{\frac{\text{Sr}}{\text{Sr} + \text{Ca}} \text{ of precursor}} \quad (1)$$

since the [Sr] is negligible compared to calcium normally present the equation becomes

$$\text{OR}_{\text{sample/precursor}} = \frac{\frac{\text{Sr}}{\text{Ca}} \text{ of sample}}{\frac{\text{Sr}}{\text{Ca}} \text{ of precursor}} \quad (2)$$

Further equations have been derived to evaluate the contribution of individual physiological processes to the over-all differential behavior of strontium and calcium. The term "discrimination factor" suitably qualified has been applied to these individual processes. In

¹ The term precursor represents the materials from which all of the strontium-calcium of the sample is derived, e.g., OR body/diet. It is especially important that the strontium-calcium ratios as measured truly represent the ratio as actually available to the organism, and as actually derived from the measured precursor.

other words a discrimination factor is equal to the OR for a simple barrier, but otherwise the OR is the product of the discrimination factors operative. Discrimination factors are important in gaining understanding of differential movement but since they are not important for present purposes, they are not discussed further in this report.

The usefulness of the OR value depends upon its constancy. Examination of OR body/diet values obtained in different laboratories by different experimental methods with laboratory and domestic animals indicate that under usual dietary conditions they range from 0.18 to 0.35, most of the values falling at about 0.25.

The OR body/diet values obtained for man call for special comment. Obviously it has been impossible to do careful studies on a sufficient number of normal individuals as has been done with animals. Examination of the work that has been done indicates a value of 0.25 from numerous stable strontium-calcium estimations on normal individuals in the United Kingdom. Limited data from the United States and Canada suggest a range of 0.25 to 0.5. Incomplete information from Japan indicates an OR body/diet value for man of 0.65; further experimentation however is necessary to establish the OR for humans under different dietary and environmental conditions especially in areas where the calcium sources consist mainly of cereals and food other than milk.

The OR for animals is reasonably constant with ordinary diets. In rats marked departure from the ordinary diet such as the presence of large quantities of milk, lactose or lysine and nutritionally abnormal Ca/P ratios, has been shown to alter the OR (34, 35, 44). It has also been observed that preweanling rats had a higher OR than mature rats.

The OR milk/diet for domestic lactating animals is an important value. The reported values for this OR for dairy cows and goats, under normal management, in different parts of the world, range from 0.09 to 0.16. Most of the values lie close to 0.11.

Application of the OR concept

STEADY STATE CONDITION

It is advantageous to be able to predict the Sr-90 body burden of an animal or human after consumption of food containing a known quantity of Sr-90. It is also desirable to be able to predict the Sr-90 content of food from the Sr-90 content of the tissues, excretions or

secretions of an animal which has consumed that food. If the OR is a reasonably constant value, such predictions can be made where the animal or human is in steady state with its nutritional environment with respect to Sr-90 and calcium. The prediction can be made by using the equation, $\text{Sr-90/Ca body} = \text{OR} \times \text{Sr-90/Ca food}$, and knowledge of the calcium content of the body and food.

NONSTEADY STATE CONDITION

In the absence of a steady state which is and will be more likely the situation, the above reasoning does not hold.

The Sr-90/Ca in the mineral being deposited in the body is equivalent to the $\text{OR} \times \text{Sr-90/Ca food}$. This mineral is being mixed with existing bone mineral and is also being deposited as new bone, especially in the young animal. Therefore the Sr-90/Ca of the total skeleton is less than the $(\text{OR} \times \text{Sr-90/Ca})$ of the food. The Sr-90/Ca of the newly deposited bone, however, approaches the value of $(\text{OR} \times \text{Sr-90/Ca})$ food. A practical aspect to be remembered is that values for Sr-90 in adult bone do not represent concentrations in local areas of bone.

A knowledge of Sr-90/Ca of food enables one to approximate the Sr-90/Ca of newly deposited bone and to predict the maximum Sr-90/Ca of any single mineral deposit in the body.

That maximum value would be equivalent to the maximum $(\text{OR} \times \text{Sr-90/Ca})$ of the food consumed.

With proper knowledge of present or predicted situations it becomes feasible to examine total diets of man or animals in regard to their contribution of Sr-90 to the population in terms of calcium supply and strontium-calcium ratios of Sr-90/Ca in individual food items.

Cesium

Radiocesium may be important as a contaminant of meat and milk of domestic animals. This element is efficiently absorbed, secreted into milk and into the cells of the body, being physiologically similar in behavior to potassium (15). For cesium-potassium, however, there is apparently not the close relationship that has been observed

for the strontium-calcium system. Much more data are required for an understanding of cesium behavior and the designation and interrelationship of important parameters. Until data are available that justify consideration of cesium in terms of potassium it is suggested that cesium levels be expressed in terms of concentrations per unit of biological material; it must be added, however, that data on potassium obtained on identical samples might well be reported for purposes of cross-checking. The advantages in consideration of cesium-potassium behavior may be listed as follows:

- (a) Natural radioactive potassium is convenient as an internal radioassay standard and may eliminate uncertainties due to differences in fat content between individuals.
- (b) The two elements are almost equally and well absorbed from the gastrointestinal tract.
- (c) The two elements tend to be concentrated within cells.
- (d) The potassium content of individuals tends to be independent of potassium intake under normal conditions.

Perhaps a major disadvantage is that cesium behavior appears to be independent of potassium levels (11).

Preliminary experiments indicate that about 6 to 10 percent of the ingested radiocesium appears in the milk of the dairy cow and goat. Survey studies show that, as of 1956-1957 in the United States, about 60 percent of the Cs-137 of the average diet is derived from dairy products, 25 percent from meat products and the remainder from vegetables, cereals and fruits.

Iodine

The presence of radioiodine in the milk of grazing animals may be most important at short times after the production and dispersal of radioactive material. A great deal is known about the various steps of iodine metabolism from the rapid absorption from the gastrointestinal tract, the collection in the thyroid gland and secretion into milk. On the average, it appears that 5 to 10 percent of the I-131 ingested as solution or from deposition on herbage by the

dairy cow appears in the milk produced, whereas values of 20-50 percent have been reported for the goat and sheep. The radioiodine appears in the milk at about 30 minutes after ingestion and decreases with a half-period of about 1.5 days for the first few days after peaking and after that with a half-period of about 3 days. Further study is needed to obtain more data especially on a continuous ingestion basis. Under normal conditions, a very high percentage of the iodine secreted into milk is in readily exchangeable form. There appears to be considerable variation between individuals in the amounts secreted into milk. Some of the factors considered as responsible include seasonal variations, feeding practices, stage of lactation and status of the thyroid gland.

Limited experiments with milk from dairy cattle fed I-131 indicate that about 0.01 percent and 0.26 percent respectively of the ingested radioactivity was found in the butter and buttermilk respectively (23).

Uptake of ingested iodine by the thyroid gland may be of some interest. In general, it appears that ingested radioiodine reaches a maximum of about 20 percent in the gland at 3 to 5 days after ingestion and thereafter is released at an apparent constant rate of about 3 percent per day. The effective half-time for the thyroid removal from cows and goats appears to be about 5 to 7 days.

Other radionuclides

It would seem prudent that experimental data in domestic animals be obtained with other radionuclides that may contaminate the food chain.

Some knowledge is already available for radioisotopes of those elements that are of physiological interest, namely Zn-65, Co-60 and radioiron. Little or no data are available in domestic animals for the other radionuclides.

Radionuclides that are poorly absorbed from the gastro-intestinal tract and do not reach the edible products of animals are of minor concern. These include Pm-147, Zr-95, Nb-95, Ce-141 and the other rare earth elements and plutonium.

Brief mention may be made of the metabolism of other radionuclides. Te-132 in a single dose was absorbed by a cow to the extent

of about 20 percent with about 0.5 percent being secreted into the milk. About 5 percent of ingested Ba-140 was found to be absorbed by the cow with about 0.4 percent being secreted into milk. Little or no data are available on the metabolism of ruthenium in domestic animals; it appears from laboratory animals that gastro-intestinal absorption is about 3 to 5 percent.

Even for such radionuclides as Zn-65, Co-60 and Fe-55 and 59, there are not enough data to permit evaluation of environmental contamination conditions. Only small amounts of these so-called micro-nutrients appear in milk and some very tentative estimates can be made from normal dietary and milk levels of these nutrients. For example, it has been calculated that for zinc, cobalt and iron about 5 percent, 0.1 percent and 0.1 percent respectively of the amount eaten daily appears in the daily milk of the cow. It may be of interest to note that milk levels of iron are not increased by increase in dietary levels, whereas substantial increases can be achieved by such means for zinc, cobalt and iodine.

In regard to C-14, it is pointed out only that the specific activity in the food chain should probably lag by less than a year that of the C-14 that is used by plants for photosynthesis (2)

Obviously, further direct evidence is needed on the metabolism in domestic animals of any radionuclides that may possibly become of importance from the standpoint of food-chain contamination.

Remedial measures

Any treatment of the animal, its environment, or animal products specifically for the purpose of reducing the radionuclide content of the human body is considered as a remedial measure. It is emphasized that such measures would be foolhardy and wasteful unless health authorities considered the levels to be present in quantities that are hazardous to health and well-being.

GENERAL

The farmer should realize that fallout contamination is a surface phenomenon; knowing this, he can protect his livestock if necessary (particularly dairy cows) by providing whatever shelter he can for animals, feed and water supplies, and restriction of grazing.

PROCEDURES WITH ANIMALS AND MAN

Practical steps to reduce the Sr-90 body burden of radioactivity in the population by procedures with man and animals would require considerable research along nutritional and physiological lines. The following are suggested as possible approaches for further work.

- (a) Consideration could be given to the use of uncontaminated calcium to contribute a maximum proportion of the dietary calcium of animals and man.
- (b) As an extension of (a) above, consideration could be given to the supplementation of diets with uncontaminated calcium. Experiments with laboratory animals and preliminary data with lactating cows and goats have indicated that under proper conditions increases of uncontaminated dietary calcium will lead to proportionate decreases in the Sr-90/Ca ratios in bone or milk (11). It must be emphasized that changes of only about two- to three-fold can be accomplished because of the abnormality of diets beyond that range.
- (c) Only small effects on the I-131 content of milk can be brought about by changes in the stable iodine content of the ration (24).
- (d) Small increases in excretion of cesium have been brought about by feeding livestock with diets that contained substances such as alfalfa, vermiculite, bentonite, oat hulls, soybean oil meal, that tended to absorb radiocesium. Little or no change in cesium levels has been produced by alterations in potassium or sodium levels.

TREATMENT OF PRODUCT

Removal of radiostrontium and radiocesium from milk

The objectives of any method of decontamination of a food is the complete removal of radionuclides without altering the composition or flavor of the food. In addition, the method should be simple to apply and relatively inexpensive.

Removal of Sr-90 and Cs-137 from milk can be achieved by treating the milk with cation exchange resins that are saturated with non-radioactive cations.

Calcium saturated cation exchange resin will remove approximately 50 percent of the strontium that enters milk via the milk secretion mechanism. If, on the other hand, strontium is added to milk in the laboratory then the same process will remove 98 percent of the radionuclide. The use of a calcium saturated resin results in a change in the concentration of Ca, K and Na in the milk.

A cation exchange resin which has been equilibrated with a solution of CaCl_2 , KCl and NaCl in which the relative proportions of Ca, K and Na are the same as in milk, removes 55 percent of naturally secreted strontium with one batch treatment and 66 percent with two treatments. This type of resin does not alter the cation composition of milk.

In order to achieve wide efficient removal, a resin must be employed which will remove a part of the calcium at the same time. Such a resin is prepared by equilibrating with a solution of CaCl_2 , KCl, NaCl, and MgCl_2 in which the relative proportions of the cations in milliequivalents are 2Ca:4K:0.5Na:0.8Mg. The proportion of these cations in milk are: 6Ca:4K:2.5Na:0.8Mg. Treatment of the milk (1 gram/20 milliliter) with this resin results in a removal of 76 percent of the naturally occurring strontium but changes the content of Ca, K and Na. Further removal to 87 percent is achieved by a second batch treatment with calcium saturated resin (0.5 gram/20 milliliter). This second treatment restores the cation composition of the milk to approximately the original state.

Cesium is removed by the same treatment to the extent of approximately 85 percent.

Yttrium is not removed by the resin treatment.

Iodine can be removed to the extent of 98 percent by treating milk, naturally contaminated, with an anion exchange resin in the chloride form. The iodine removal experiments were conducted in the United Kingdom, using the column technique.

None of these removal methods has reached the pilot plant stage.

Removal of radiocesium from meat

Preliminary experiments on small samples from laboratory animals suggest that the removal of Cs 137 from meat may be possible and merit further investigation.

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RADIOACTIVE CONTAMINATION IN FISHERIES PRODUCTS

Introduction

The purposes of the report are:

1. to indicate the present level of contamination of the sea and of freshwater bodies, and of the fisheries products from each of these environments, resulting from flow-out¹ and fallout of fission products from weapons tests;
2. to indicate the contribution to the human diet of radionuclides originating in fisheries products;
3. to estimate the probable future contamination of fisheries products from flow-out and fallout;
4. to evaluate existing and potential sources of radioactive materials to the freshwater and marine environment originating from the peaceful uses of nuclear power;
5. to present references to published material which give general approaches to the problem of evaluation of the capacity of the marine environment to receive radioactive materials;
6. to indicate areas in which present knowledge is lacking, and to recommend research required to supply this required knowledge.

It is only in a few countries that fisheries products contribute more than a few percent of the food intake for the human population. Partly for this reason, and partly because of the low level of contamination of fisheries products compared to some agricultural pro-

¹ In addition to direct fallout onto the sea and other natural water bodies, radioactive materials are also washed off from land into the freshwater body or into the sea. Such addition of radioactive materials to freshwater and marine environments is here termed *flow-out*.

ducts, the same attention has not been given to the contamination of fisheries products from radionuclides derived from weapons testing as there has been for food products originating on land. Japanese efforts represent an exception to this statement. As a result of the limited amount of data available, it has not been possible to treat the probable latitudinal differences in fallout, and only relatively broad statements as to the degree of contamination in the Pacific Ocean on the one hand, and the North Atlantic Ocean on the other hand, are made here. Certain specific data on the concentration of various isotopes in the marine environment, and in marine and fresh-water fisheries products, are quoted in this report. Because of the limited nature of the data it was not possible critically to evaluate the validity of the values given, and such data cannot be utilized for rigid statistical or numerical treatments.

Natural radioactive background in the sea

In order to have a proper base upon which the discussion of the effects of artificially produced radionuclides in the sea may be made, the amounts and kinds of naturally occurring radioisotopes in the environment should be known. In Table 5 a list of natural occurring radionuclides, together with the amount of the isotope, in tons and in curies, and the amount of the total element, both active and inactive isotopes (1).

TABLE 5. — RADIOACTIVE ELEMENTS IN SEA WATER

Element	Concentration, mg/kg	Total in oceans (tons)	Natural activities			
			Nuclide	Tons	Curies	Conc. $\mu\text{c./liter}$
Potassium	380	5.3×10^{14}	K-40	6.3×10^{10}	4.6×10^{11}	330
Carbon	28	3.9×10^{13}	C-14	56	2.7×10^8	0.19
Rubidium	0.2	2.8×10^{11}	Rb-87	1.18×10^{11}	8.4×10^9	6.0
Uranium	0.0015	2.1×10^9	U-238	2.8×10^9	3.8×10^9	2.7
			U-235	2.1×10^7	1.1×10^8	0.08
			Th-232	1.4×10^7	8×10^6	5.7×10^{-1}
Radium	$0.2 \cdot 3 \times 10^{-10}$	28-420	Ra-226	4.2×10^2	1.1×10^9	0.27

These naturally occurring radioisotopes produce a beta activity of approximately 340 $\mu\text{uc/liter}$, about 90 percent of which results from K-40.

The incident cosmic radiation of about 35 mrad on the sea surface is rapidly absorbed by the sea, such that only about 30 percent reaches a depth of 10 meters and 4 percent reaches a depth of 50 meters.

The radioactivity of the sea bed varies with the type of sediment. Highest activities are found in deep water sediments, of which red clay has a higher activity than blue clays and globigerina oozes. The radioactivity of shallow water sediments with which commercial marine food resources may come into contact, have considerably lower natural radioactivity than the average soil on land. It is about one third of the natural radioactivity of granite rocks.

Marine animals living in the sea also receive, besides the external radiation described above, some radiation from internal sources, mainly from radiopotassium accumulated in the tissues and bones. The differences between the radiations received by man and large fish in the sea is obvious from the fact that at the surface of the sea the natural level of radiation in fish amounts to about two thirds of the level of man over land; this figure decreases to about one half at 8 meters depth and to about one third at 30 meters depth. The average natural radiation received by man is of the order of 100 mrad per year.

The naturally occurring radioactivity in freshwater environments is highly variable, dependent on the type of geological structure being drained to supply the fresh water. No general statements such as those made for sea water can be given in the case of fresh water.

Effects of weapons tests

In this section, the effects of direct fallout of radioactive materials on the sea and on freshwater environments, as well as the flow-out of such material from land to the sea and into freshwater bodies, will be discussed.

ACTIVITY IN THE SURFACE LAYERS OF THE OCEAN DUE TO WEAPONS TESTS

In an oral presentation made at the International Congress of Oceanography, held in New York City in September, 1959, the Russian oceanographer, B.A. Nelepo, reported that measurements made in 1958 with an *in situ* beta detector showed activities in the upper 100 meters of the South Pacific Ocean of over 700 $\mu\mu\text{c}$ /liter, or somewhat more than twice the natural background. Below 100 meters the activity returned to levels near the natural activity due to K-40. No measurements of specific isotopes were made, and much of this activity is probably due to short-lived fission products. It is evident that very little of this relatively large increase in activity above natural background can be attributed to Sr-90. The Japanese have found a Sr-90 concentration in the surface layers of the Pacific of about 1 $\mu\mu\text{c}$ /liter. If we assume that surface fallout over mid-latitudes of the ocean areas of the world up to mid-1958 had been about 10 millicuries/square kilometer for Sr-90, the concentration of activity in a mixed surface layer of 100 meters' depth should be about 0.1 $\mu\mu\text{c}$ liter from Sr-90. Measurements by Bowen (2) in the surface layers of the Atlantic near the East Coast of the United States are reasonably consistent with this value of 0.1 $\mu\mu\text{c}$ /liter. The highest Sr-90 in the Pacific evidently results from the dispersion throughout much of the Northern Pacific of the effects of close in fallout and underwater shots in the Pacific test area.

Bowen (2) has also made Ce-144 measurements. These figures are of the order of five times the activity due to Sr-90. Now the MCP value (3) for drinking water for Sr-90 is about 1/100 that of Ce-144. However, the concentration factor² for Ce-144, for edible parts of many invertebrates in the sea, is over 100 times that for Sr-90, and for the soft parts of fish perhaps 10 times that of Sr-90. Thus, Ce-144 appears to be as important, and perhaps more important from the standpoint of certain sea foods, as Sr-90, in regard to recent fallout. However, the concentration of activity in sea food for both of these isotopes is probably rather insignificant compared to the contribution from terrestrial food sources.

² Concentration factor equals the ratio of the amount of the element per unit weight of the organism to the amount of the element per unit weight of sea water or fresh water.

ACTIVITY IN MARINE FISH RESULTING FROM WEAPONS TESTS

Hiyama (4) found that the surface fish have a Sr-90 concentration which, when taking into account species differences, is in equilibrium with the $1 \mu\mu\text{c/liter}$ concentration for the surface layers of the Pacific, or about $0.1 \mu\mu\text{c/kilogram}$ of fish flesh and $0.3 \mu\mu\text{c/kilogram}$ of whole fish. However, fish which live below 100 meters in depth have Sr-90 concentrations which appear to be in equilibrium with environmental concentrations of 10^{-1} to $10^{-2} \mu\mu\text{c/liter}$.

The concentration of calcium in sea water is quite constant at close to 400 milligrams/liter. In the North Pacific, the ratio of Sr-90 to calcium in the surface layers is then $2.5 \mu\mu\text{c Sr-90/gram Ca}$. The corresponding figure for the North Atlantic would appear to be of the order of one tenth this value. There appears to be a discrimination factor of about seven favoring calcium against strontium in fish. The considerations lead to the conclusion that the Sr-90/calcium ratio in fish in the North Pacific surface layers was then, in 1958, about $0.3 \mu\mu\text{c Sr-90/gram Ca}$. This value is used in the next section to determine an upper limit for the probable contribution of Sr-90 from sea food to the human diet.

Donaldson (5) detected Ce-144 plus Pr-144 in fish caught in 1959, and Saiki (6), Amano (7), Kawabata (8) found that Zn-65, Fe-55, and Fe-59 were the main radionuclides in the soft tissues of fish caught in the Pacific in 1954. Saiki (9, 10, 11) determined the amounts of radionuclides contained in marine fishes caught in the Pacific Ocean from 1954 to 1958. The concentration of Zn-65 of highly contaminated fishes in 1954 were as follows: $10^1 \mu\mu\text{c/gram}$ in the liver, kidney and spleen; $10^2 \mu\mu\text{c/gram}$ in red muscle; $10^2 \mu\mu\text{c/gram}$ in ordinary muscle. The concentration of Zn-65 decreased in 1956, being less than $10^2 \mu\mu\text{c/gram}$ in the intestinal organs. In fish caught in 1956, some Cd-113m, Cd-115m, and very small amounts of Mn-54, Sr-90 plus Y-90, and Ba-140 plus La-140 were found together with the previously identified nuclides such as Zn-65, Fe-55 and Fe-59. For example, the analysis showed that the liver of a highly contaminated big-eyed tuna *Parathunnus mebachi*, caught in October 1956 had the following concentrations of radionuclides: $5 \times 10^3 \mu\mu\text{c/gram}$ of Fe-55; $5 \times 10^2 \mu\mu\text{c/gram}$ of Zn-65; $1.2 \times 10^2 \mu\mu\text{c/gram}$ of Cd-115m; $22 \mu\mu\text{c/gram}$ of Mn-54; $20 \mu\mu\text{c/gram}$ of Sr-90; and traces of Ba-140 plus La-140. The concentrations of Fe-55 in the intestinal

organs were 10^3 to 10^4 $\mu\mu\text{c}/\text{gram}$ in 1956. In an analysis of big-eyed tuna caught in December 1957, one year after previous weapons tests, only Fe-55 (13.3×10^4 $\mu\mu\text{c}/\text{gram}$ and 1.4×10^1 $\mu\mu\text{c}/\text{gram}$ in the red muscle and ordinary muscle, respectively) was found. Welander (12) also found Zn-65 (of the order of 10 $\mu\mu\text{c}/\text{gram}$) Co-57, Co-58 and Co-60 (of the order of 6 $\mu\mu\text{c}/\text{gram}$), Fe-55 (approximately 6 $\mu\mu\text{c}/\text{gram}$) and smaller amounts of Mn-54 and Sr-90 (about 1 $\mu\mu\text{c}/\text{gram}$ for the latter isotope) from fish collected on reefs in Rongelap Atoll in 1957.

These data are of value in indicating the pattern of distribution of radionuclides in fish, but all the quoted authors have pointed out that these analyses had been done on particularly highly contaminated fish, and do not represent the levels of contamination in fisheries products in general, even from the Pacific.

On the basis of intercomparisons of various isotope concentrations, it appears that the specific specimens discussed above were contaminated by factors of from 100 to 1,000 over that which might now be expected for surface fish in general in the North Pacific. The concentrations of Zn-65 and Fe-55 in fish in the surface layers in the Pacific were probably then of the order of 0.01 $\mu\mu\text{c}/\text{gram}$ and 0.1 $\mu\mu\text{c}/\text{gram}$ respectively. It might be noted that the concentration of inactive zinc in the ocean is about 5 μ gram/kilogram, and that of iron varied from 2 to 20 μ gram/kilogram. It should be noted that the iron in sea water occurs in both dissolved form and as a colloid, and the amount available for biological processes is not known.

Hiyama (13) also gives analysis for marine fish of Cs-137 from more representative specimens and indicates a concentration of about 0.02 $\mu\mu\text{c}/\text{gram}$.

Contribution to the human diet of radioactive isotopes in sea food

Hiyama (13) quotes a diet for urban inhabitants of Japan which indicates the daily intake of sea food by this population group as about 100 grams per person, involving about 100 milligrams of calcium per person per day from this source. This diet includes a mixture of small fish in which case bones as well as flesh are eaten, and large fish from which only flesh is eaten. The concentration of 0.3

$\mu\mu\text{c}$ Sr-90/gram Ca which appears probable for fish from the surface layers of the Pacific would then provide about 0.03 $\mu\mu\text{c}$ Sr-90/person/day, to the diet of this urban population in Japan. The corresponding daily intake per person of Cs-137, Zn-65 and Fe-55, on the basis of assumed concentrations of 0.02 $\mu\mu\text{c}$ /gram of edible fish, of 0.01 $\mu\mu\text{c}$ /gram edible fish, and 0.1 $\mu\mu\text{c}$ /gram edible fish respectively, is then 2.0 $\mu\mu\text{c}$ Cs-137, 1.0 $\mu\mu\text{c}$ Zn-65 and 10 $\mu\mu\text{c}$ Fe-55.

In view of the fact that the MPC values (3) for drinking water for Cs-137, Zn-65 and Fe-55 are higher than for Sr-90 by factors of 200, 1,000 and 8,000 respectively, these isotopes are evidently much less important than Sr-90 insofar as human intake of sea food is concerned, at least with respect to nuclides originating from weapons testing.

Other ocean areas are probably much less contaminated than the Pacific and few populations consume as much as 100 grams of sea food per person per day. Hence the case of the urban inhabitants of Japan may be taken as representing a population group in which the fraction of radioactive materials in the diet which originates from the sea is near the maximum value. Even in this case the contribution from the sea is quite small. The present intake of Sr-90 is estimated at from 6 to 10 $\mu\mu\text{c}$ person/day. The sea then contributes at most between 1/200th and 1/300th the total daily intake of Sr-90.

Contribution to the human diet of radioactive isotopes in freshwater fish

The concentration of radioactivity resulting from weapons tests in freshwater bodies is highly variable, depending on the complex processes of washoff from adjacent land areas, geochemical processes of sorption and exchange involving suspended sediment and bottom materials, and biological uptake. There are insufficient observations to allow any generalized statements to be made regarding the concentration of the various radionuclides in freshwater bodies.

The only measurement which appear useful for this analysis are those made in Japan. Hiyama (13) indicates that the ratio of Sr-90 to calcium in freshwater fish was about 20 times that in marine fish. Thus a population group which consumed 100 grams per person per day of freshwater fish, involving about 100 milligrams of calcium per person per day (includes some fish bone intake), would receive

about 20 times as much Sr-90 from this source as a population group eating the same amount of marine fish would receive from that source. However, the contribution of Sr-90 to the human diet of a population group eating freshwater fish would still be a small fraction of the total Sr-90 intake from all foods, being only about one tenth to one hundredth the total intake.

Effects of peaceful uses of nuclear energy

The discussion above has indicated that present contamination of fisheries products from fallout is small compared to other food products. It should not be concluded from this evidence, however, that there is no necessity to continue investigations concerned with the fate of radioactive materials which have been or might be introduced into the sea or freshwater environments. For one thing, it is possible that the contamination of marine products will not decrease, after cessation of weapons tests, at the same rate as for land food products, and hence the contribution of radionuclides from fisheries products may become relatively more important, in comparison to the contribution from land food sources, with time. A factor of even greater importance, at least for certain marine and freshwater locales, may be the effects of the peaceful uses of nuclear energy. An unavoidable consequence of the utilization of atomic energy for power production and other peaceful purposes is the production of unwanted fission product wastes. While over 99.9 percent of these wastes are now and will probably continue to be contained and stored on land, it is not practical to avoid completely the release of some activity to the environment. Such materials now enter and will without doubt continue to enter the atmosphere; to fall out on land; to be introduced into freshwater systems; and to be introduced into the sea. The total amount of activity which enters the environment as a result of peaceful uses of atomic energy is very small compared to that resulting from weapons tests, and the rate of introduction to all parts of man's environment is likely to remain only a fraction of the recent rate of introduction to such environments from weapons tests. However, with the increased utilization of nuclear power, it is quite likely that local effects of probable releases of activity to the environment will become quite significant.

There are several different groups of sources of artificial radiation to the marine and freshwater environments. These sources produce radioactive wastes with very different composition and level of activity.

One treatment and refining produces low level wastes which contain mainly uranium and daughter products.

The vast majority of the fission materials produced in a reactor remain contained in the fuel elements while such elements remain in the reactor. In the case of gas-cooled reactors radioactive wastes could enter the aquatic environment only through fallout from materials released to the atmosphere, possibly from leakage through the heat exchanger systems to the condensor water, or leakage from spent fuel element storage pits, all unlikely routes. In pressurized water reactors leakage from the primary system and from spent fuel elements storage pits might contribute radioactive materials to the aquatic environment.

Large research and experimental atomic energy facilities may provide a larger source of radioactive materials which may be released to the environment than an operational power producing reactor. The amount and nature of such materials would be quite variable because of the experimental nature of the work being conducted at such facilities.

The chemical processing of spent fuel elements provide the largest source of radioactive substances which are now being released to the environment. After separation and storage in tanks of the vast majority of the fission products, a small fraction of the wastes remain in a high volume liquid effluent which in some cases is released to the marine environment. Induced activity in any circulating water for fuel element storage pits also leads to activity being introduced, in some cases, to freshwater bodies.

The use of radioactive isotopes in various fields of scientific research as well as in medicine produces liquid and solid radioactive wastes of predominantly medium and low activity. Low activity effluent wastes are usually disposed of, after dilution, in sewers, and the solid wastes are packaged and disposed of in land areas or in the sea. The total amount of activity from these sources is very low.

It seems unlikely that the release of some of the activity produced by nuclear powered ships into the sea can be avoided. In presently designed water moderated reactors for ships, operational wastes,

contaminated principally by induced activity of corrosion products will result from leakage from the primary coolant loop, from laboratory and laundry wastes, and from operational decontamination of personnel and equipment. Whenever the reactor is warmed up after a stay in port, a part of the primary coolant is displaced. Such warm-up volumes might, after limited storage, be released to the sea. The major potential source of operational wastes from a nuclear powered ship which might be released to the environment are the spent ion exchangers used in the clean-up loop of the primary coolant system.

Information has been published regarding the introduction of radioactive wastes from a chemical processing plant into the marine environment (14). Studies regarding disposal of packaged low level radioactive wastes into the sea, and of the problem of radioactive wastes from nuclear powered ships, have also been published (15 and 16).

Present status of knowledge of the physical, chemical, geological, and biological processes in the marine and freshwater environments pertinent to the problem of radioactive contamination of fisheries products

There are a number of excellent published documents which cover much of the material for this section. No attempt will be made here to repeat the extensive material in these documents. Instead, appropriate references will be given.

A comprehensive review of our present knowledge (up to 1957) of the processes in the sea pertinent to the radioactive contamination of that environment, and of the products harvested therefrom was recently published (1). Further information on our present knowledge of the pertinent processes is given (15 and 16) which also show how this existing knowledge can be used to evaluate specific problems of introduction of radioactive materials to the marine environment. Pritchard (17) also gives a general approach to the evaluation of the safe capacity of the sea to receive radioactive materials.

Quite valuable information on the physical, chemical and biological processes operating in coastal waters can be extracted from the published reports on the preliminary studies and operational surveys made in connection with the Windscale works of the United Kingdom Atomic Energy Authority.

A number of papers have been published in the scientific literature dealing with laboratory measurements of uptake of specific radionuclides by certain marine and freshwater organisms. For a representative bibliography in this field see (18). Japanese studies of the distribution of radioactivity in the Pacific resulting from weapons tests, and their reports on the concentration of activity in fish and other marine organisms, also serve as valuable sources of information on the various processes in the sea pertinent to the problem of introduction of radioactive materials into the sea.

A review of the available published material indicates certain important gaps in our present knowledge. The present extensive studies of biological uptake of radioactive isotopes deal primarily with single species and provide information on only individual steps in the movement of material through the various trophic levels of the food chain. No published reports are available on any comprehensive study of the movement of radioactive substance through the entire food chain. Also no data are available for certain isotopes, such as Ta-182, which appear to be important corrosion products in some pressurized water reactor wastes.

Current available information, derived primarily from word of mouth discussions of research projects now in progress, indicate the concentrations of activity which would be required to disrupt the natural ecosystems³ to any measurable extent are considerably higher than any concentration which could be tolerated from the standpoint of safety to man's utilization of the products from the sea. Hence it would appear that if radioactivity in the sea does not reach levels which would limit man's use of products from this environment, then there would be no danger to the marine biota from radiation damage. However, documented evidence is required in this important area.

Physical processes of importance in the evaluation of the fate of radioactive materials introduced into the sea, include dispersion within a given segment of the sea and exchange between the various segments of the sea. These are areas of active oceanographic research and progress is evident from recent publications and oral presenta-

³ *Ecosystem* (ecological system): A system comprising the plant and animal communities (biota) and their environment, including the interactions between the biota and the environment.

tions at scientific meetings. However, attainment of adequate knowledge in these areas required continued support and perhaps intensification of effort. The question of the residence time for the deep layers of the sea, and the exchange between the productive surface layers and the deep layers, is particularly pertinent. The role that living organisms play in this exchange is not known, and research on this subject is desirable.

An important geochemical problem with respect to the ultimate availability of radioactive materials to the food chain involves the rate of movement of various elements from solution or colloidal suspension in the sea to incorporation into the bottom sediments. While the work of Goldberg and Arrhenius (19) is an important contribution in this regard, no published information exists for a number of elements which appear as radioactive corrosion products in pressurized water reactors.

Conclusions

In the previous section a number of areas in the fields of fisheries biology and oceanography, in which information pertinent to the possible radioactive contamination of fisheries products appears particularly weak, were discussed. Actually, additional knowledge in many other areas in these fields would be useful in regard to the subject under consideration in this report. Rather broad conclusions regarding the need for increased research efforts in freshwater and marine biology and in oceanography could be made, and in fact have been made, by several national and international groups. This Committee considers that conclusions presented here should be specifically directed towards those areas of concern to FAO.

SPECIFIC AREAS OF RESEARCH ON MARINE AND FRESHWATER ECOSYSTEMS IN WHICH SUPPORT OR ENCOURAGEMENT BY FAO IS DESIRABLE

It appears appropriate that this Organization concern itself primarily with promoting research in areas of freshwater and marine biology in which knowledge is particularly important to the question of radioactive contamination of fisheries products. A second general area, in which action appears appropriate for this Organization,

involves human nutrition -- in particular with respect to the subject matter of this appendix, the relative contribution from fisheries products of specific elements to the human diet.

In the field of aquatic biology, both freshwater and marine, research on the transport of radionuclides through the complete aquatic food chain under natural conditions should be encouraged. The major portion of the studies on uptake of radionuclides in the marine and fresh water biota (total living mass) which have been undertaken to date involve individual species and frequently laboratory conditions not representative of the actual natural environments. The complex interaction of the several trophic levels have not been adequately included in these studies. Some studies have indicated that the final equilibrium level of activity in the organisms of the higher trophic levels is not dependent on route: that is, the concentration of specific radioactive isotopes finally reached in fish is the same whether the uptake is directly from the water or from the ingested food. However, the rate of uptake does appear to be dependent on the route the elements take in reaching the fish. However, evidence is limited on this important subject and further research should be encouraged.

The present operations of the Windscale Works of the United Kingdom Atomic Energy Authority, in which certain quantities of radioactive waste materials are introduced into the coastal waters of the Irish Sea, offers an excellent opportunity for the study of the physical, chemical, and biological processes involved in the ultimate transport of specific radionuclides through the marine food chain under natural conditions. It is evident from the published data on the operations at this site that the public health is adequately safeguarded, and that the current monitoring program ensures the continuation of such safeguards. Added intensive scientific studies, involving organisms not now utilized by man and involving levels of activity below those important from the standpoint of human intake, would contribute a very significant fund of knowledge upon which an understanding of the natural physical, chemical, and biological processes involved in the transport of radioactive substances in the aquatic food chain could be based. FAO should encourage the continuation and intensification of such studies, and in particular promote the dissemination of the results to interested research activities throughout the world.

In the area of nutrition the trace element concentration in the human body, in the human diet, and particularly in the marine products contributing to the human diet should be emphasized. Of the radionuclides arising from peaceful uses of nuclear energy many are isotopes of trace elements, such as cobalt, copper, iron, etc., which enter certain biochemical processes.

CO-OPERATION AND CO-ORDINATION AMONG INTERNATIONAL ORGANIZATIONS

A number of international organizations, including the Food and Agriculture Organization (FAO), the International Atomic Energy Agency (IAEA), the World Health Organization (WHO) and the United Nations Educational, Scientific and Cultural Organization (UNESCO), have responsibilities which involve directly or indirectly the subject of radioactive contamination of fisheries products.

In particular, there exists close co-operation between FAO and the IAEA in the field of radioactive waste disposal to the sea. This co-operation includes the active participation of the Fisheries Division of FAO on the IAEA Expert Panel on the Disposal of Radioactive Wastes in the Sea, and of IAEA on this Committee. Strengthening of this existing co-operation and co-ordination might involve joint sponsorship of expert panels and of scientific meetings.

It is recognized that FAO is not in a position to support financially research efforts by individual scientists or laboratories in this field. In view of the fact that IAEA does have funds designated for support of research, it would be desirable to have the existing co-operation between FAO and IAEA in sponsoring research in the field of radioactive waste disposal to the sea strengthened. The Fisheries Division of FAO has the required background upon which to base recommendations regarding support of particular research projects in aquatic biology which would contribute to required knowledge in this field. Some formal mechanism might be desirable to replace the present form of contact between FAO and the IAEA regarding sponsorship of research in this important area.

Consideration should be given to joint planning by FAO and IAEA for the collection and assay of individual nuclides of samples from the aquatic environment in regard to radioactive contamination.

The expert knowledge of the Fisheries Division of FAO could then be well combined with the facilities of IAEA.

Consideration should also be given to joint efforts by FAO and IAEA to institute, through their Member Governments, means whereby research results in the area of radioactive contamination of aquatic environments and biota conducted at the national level could be promptly transmitted to interested research institutes in other nations of the world. In this regard, the existing bibliographic activities of the Fisheries Division, FAO, might be extended to include close contact with research groups in all nations and the dissemination of research results from each such research activity to others, perhaps through a regular abstracting service in this field.

It is desirable that FAO take an active role in encouraging the continuation of the oceanographic data centers, established during the recent International Geophysical Year. In particular, efforts should be made to have these centers include in their compilations data on radioactive concentrations in the aquatic environment and in aquatic biota. Of particular importance is the inclusion, in the operating procedures of such centers, of means for ready availability of the collected data.

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THE EFFECT OF FOOD CONSUMPTION HABITS AND PROCESSING ON THE CONTENT IN HUMAN DIETS OF SOME RADIOACTIVE AND STABLE ISOTOPES, WITH SPECIAL REFERENCE TO STRONTIUM, CALCIUM, CESIUM AND IODINE

In connection with the study of radionuclides as sources of internal radiation, it is necessary to have information on food consumption and food processing, i.e., on diets habitually consumed by human beings, for the following reasons.

- (a) Data on the consumption of foods are needed to assess the intakes of these nuclides by human beings through their diet.
- (b) Such data can be used as a guide to the development of appropriate monitoring programs in which foods are analyzed for their content of nuclides, particularly those of biological importance, viz., Sr-90, Cs-137 and I-131.
- (c) Information on food processing and preparation methods, considered along with the results of such analysis, is useful in considering ways of reducing the hazards arising from contamination of foods. Cereal milling practices are an example of this.

Food consumption data

There are two main sources of data on food consumption patterns and levels. The usual source of data on national average consumption is the food balance sheet which is merely an estimate of the *per caput* supplies of foodstuffs available for human consumption in a country or a territory as a whole. The other important source of food consumption data is the dietary survey which, however, is often limited in practice to small sections of the population, except in a few countries.

FOOD BALANCE SHEET METHOD

Consumption data through the food balance sheet method are derived by adjusting the quantities of foods produced in a country or territory to allow for imports and exports as well as changes in stocks to give gross food supplies. If from these amounts are deducted the amounts used otherwise than for human consumption, i.e., for animal feed, seed, manufacture and wastage, the remainder represents the total food supplies available for human consumption at the "retail level", that is, the level of the shopping basket (13). Food consumption levels derived in this way relate to national or territorial populations as a whole and thus conceal important differences in the consumption by different sections of the population, e.g., by regions, economic or social levels, age and occupation.

DIETARY SURVEY METHOD

Detailed information on such differences can be obtained only through surveys on representative samples, usually on a family basis, of different groups of the population, including those living in different regions of a country. These require direct investigation of food consumption. This is usually obtained from records of the quantities of foods taken into the kitchen by the housewife. Such records can provide information on the amounts of various foods consumed by families belonging to different sections of the population. It must, however, be noted that, whereas food balance sheet data are available on an annual basis for a large number of countries, family food consumption surveys have been comparatively limited both as to the number of countries undertaking them and the proportions of their populations covered (21). For such surveys, it is necessary first to select a sample of the population or group to be studied, unless the group is really small. In some countries, for example Israel, representative samples of the working population have been selected from cards kept by trade unions. In the United Kingdom, for the National Food Survey, samples of the whole population in private households have been obtained from the lists of addresses in the electoral registers. Many difficulties may arise in sampling. For example, some of the families approached may not be prepared to co-operate or having agreed to co-operate may, for reasons ranging

from sickness to inability to make accurate records, not be able to provide useful information.

In order to assess the hazards to individuals, it may be considered necessary to measure the dietary intakes of individuals but neither food balance sheets nor family food consumption surveys can provide such information on the consumption of individuals. To obtain such information on a national scale is prohibitively expensive, both in money and efforts of investigators. Further there is evidence of a tendency among some types of people to change their dietary habits when their individual diet is under investigation. Thus, it may be both impracticable and also not useful to attempt to survey the diets of individuals, as opposed to families, for the assessment of the intake of Sr-90 or any other nuclide except on a limited scale for special purposes. It may be necessary to assess intake of nuclides from dietary sources for groups of individuals with special physiological needs, such as children, pregnant women, the sick or the aged, or of individuals, such as isolated groups of workers, thought to be a special risk. In such groups *ad hoc* special surveys could be designed, though specific techniques might have to be devised for each group. In the United Kingdom, for example, these problems were considered and it was decided to assess the ratio of Sr-90 to calcium for the diet as a whole, based on the average results of the National Food Survey (10), but to pay particular attention to milk because of its importance as the sole diet of some infants. It was recognized, however, that no direct information on the diets of representative groups of infants and young children was available and that if this was needed special arrangements would have to be made to collect it.

Water consumption

It is not easy to find records of water consumption in any country. No estimates are included in food balance sheets and records are not usually collected in family food surveys. It is not uncommon, however, to record the volume of water and other beverages taken by individuals in individual dietary studies and it may be possible to make an estimate of national water consumption from such sources. This should include the consumption of soft drinks and, where

appropriate, of alcoholic drinks. An estimate, based on the results of a survey on the nonalcoholic beverage consumption of men, women and children, was used to determine the possible contributions of Sr-90 and calcium made by water in the average British diet. In contrast, the I.C.R.P. estimate of 2.2 liters per day average water intake in foods and fluids in a standard man was used in the Japanese "standard recipe" calculations made to assess the diets of rural and urban inhabitants in Japan (15).

The known variations in the concentrations of radioactive and stable isotopes in water, whether in different countries or within individual countries, make it important to attempt to obtain records of water consumption which are likely to vary greatly among individuals and among different population groups, and to include water and other beverages in analyses and estimates of contamination of foods. If it was made known to authorities conducting individual dietary surveys that such information was required, it should not be difficult to collect records of fluid consumption along with those on food consumption. It would never be easy to collect such records in the course of family food consumption surveys which, as already explained, usually relate to the quantities of food obtained by the housewife for the whole family.

Strontium-90

The dietary aspects of Sr-90, Cs-137 and I-131 are not similar. They will, therefore, be considered separately.

On the basis of present knowledge it seems most convenient to study the calcium contribution of diets in order to obtain information on Sr-90 contamination. The basis for this is discussed in Appendix 3.

It is recognized, however, that when the system is in a steady state the strontium contribution will not be directly related to the calcium levels in the diet: this is illustrated in Table 6 (22).

Under the nonsteady state condition that exists at present, a calcium assessment of the diet is helpful because, with knowledge of biological discrimination and food chain pathways, reasonable predictions can be made on the importance of foods as contributors of contemporary Sr-90 in the human population.

TABLE 6. - RATIO OF STABLE STRONTIUM TO CALCIUM IN VARIOUS TYPES OF FOOD

Food	mg Sr/g. Ca
Cereals and vegetables	2
Milk and milk products	0.3
Marine fish	3
Freshwater fish	1

Intake of calcium

When it comes to the assessment of the intakes of some nutrients, such as calcium, in contradistinction to the consumption of foods, food balance sheets are particularly unsatisfactory, since statistics on some of the main sources of calcium, viz. dairy products, fish and vegetables, are often extremely limited, both in quantity and quality. Apart from the inadequacy and inaccuracy of these statistics, the breakdown of the food groups usually is not sufficiently detailed to enable the calcium content to be calculated with any reasonable degree of assurance. For instance, there is a great difference between the calcium contents of leafy vegetables and other vegetables and between those of small fish (eaten whole with bones) and bigger fish, but available statistics do not usually differentiate between them. Further, the effects of food processing and preparation, such as the addition of calcium carbonate (*creta preparata*) to flour in the United Kingdom or the making of maize tortillas in Mexico and Central America, in which the initial stage is to treat the maize grain with lime water before grinding, cannot usually be taken into account in making estimates based on food balance sheets.

Nevertheless, in spite of these serious limitations, national food balance sheets can be used to give a rough general estimate of the calcium supplies contributed by the major food groups. Such estimates have been made using the analyses of foods for calcium given in *Food Composition Tables - Minerals and Vitamins for International Use* (11). The results of these calculations are given in general terms

in Annex A at the end of this appendix. From these it is clear that in different countries not only calcium intakes vary widely but also the contributions of different food groups.

The countries shown can be grouped into four broad categories, which include:

1. Most of Europe, North America and Oceania, with over 800 milligrams of calcium per head per day, of which 70 to 90 percent is obtained from milk and milk products.
2. Southern European countries, such as Italy and some Latin-American countries, such as Argentina and Uruguay, with 600 to 700 milligrams of calcium daily, about 60 to 80 percent of which comes from milk and milk products but for which vegetables tend to make a relatively more important contribution than in the first category. (Israel fits by pattern into this category, but has a higher total calcium consumption, mainly because of the large contributions from vegetables).
3. Egypt, Turkey, India, Pakistan and the Union of South Africa, in which milk and its products are still the main contributors (30 to 65 percent) but to a much smaller total calcium intake of 300 to 450 milligrams per head per day of which cereals, pulses and nuts and vegetables become relatively more important.
4. Chile, Ceylon and Japan, where milk and its products do not make the major calcium contribution and where the daily consumption of 200 to 350 milligrams per head is derived fairly evenly from cereals, pulses and nuts, vegetables and dairy produce. Only in Chile and Japan does fish appear to make an important contribution.

Similar impressions can be gained from a study of the results of family food consumption surveys. As the number of such surveys is small, it is not possible to classify their results in any systematic way. However, the following few interesting features are indicated by study of survey data from a few countries (*see* Annex B).

- (a) Calcium consumption appears to vary from less than 400 milligrams per head per day in parts of Africa and Asia to over 1,000 milligrams in the United States of America and the United Kingdom.

- (b) Fish and other marine foods are very important in some countries like Japan and Ivory Coast, especially among coastal people, as small fish (often eaten with bones) and shellfish, which are rich in calcium, are frequently eaten in those areas.
- (c) Cereals appear to be the most important source next to dairy produce in countries like India.

The results from dietary surveys bring out another interesting fact that the differences in calcium intakes and principal sources of calcium are significant not only between different countries but between different sections of the population within individual countries. Table 7 shows the variation in calcium intakes among different sections of the population in Ceylon (6).

TABLE 7. - AVERAGE CALCIUM INTAKES IN CEYLON (1948-1949)

Type of population	Economic level 1	Economic level 2	Economic level 3
 (mg. per day per consumption unit)		
<i>Racial origin</i>			
Sinhalese	425	600	812
Ceylon Tamil	290	410	595
Indian Tamil	341	610	1 315
Ceylon Moor	297	415	580
<i>Urban - rural</i>			
Urban	319	479	741
Rural	416	594	749
Estate	341	610	1 315

It will be seen that calcium intakes are influenced significantly by economic level, both on the basis of racial origin and of residence in town or country, but in all cases higher economic levels indicate higher calcium intakes. Moreover, within each economic level, the rural areas seem to have higher intakes than the urban areas.

Apart from these differences in total calcium intakes between

urban and rural sections of such a population, there may also be differences in the relative contributions of different food groups to the total calcium intake. Table 8 shows the results of some studies in India which illustrate this point (29).

TABLE 8. - PERCENTAGE OF CALCIUM DERIVED FROM DIFFERENT FOODS IN AVERAGE INDIAN DIETS (1935-48)

Food	Agriculturists	Industrial laborers
 Percent	
Cereals and pulses	56	39
Vegetables and fruits	22	26
Milk and milk products	12	14
Other foods	10	21

It appears that there are important differences in the contribution of some foods such as cereals and pulses whereas with others like milk and its products the differences may be negligible. A similar point, though different in detail, emerges from a study of the sources of calcium in the so-called "standard recipe diets" for rural and urban inhabitants in Japan devised by the National Institute of Nutrition (15). In the "recipe" for rural inhabitants, nearly half the calcium comes from vegetables and fruits, whereas in that for urban inhabitants milk is the chief source, supplying nearly 40 percent compared with less than 20 percent from vegetables and fruits and a similar proportion from fish.

Unusual sources of calcium

Reference has already been made to the fact that some foods, such as rice and other cereals, which are not particularly rich in calcium, can be one of the most important contributors of calcium in some diets characteristic of the underdeveloped areas. This will be especially true where the staple cereal is either ragi (*Eleusine coracana*) or quinoa (*Chenopodium quinoa*), both of which are exceptionally rich in calcium.

Among the cereals widely consumed in India, ragi -- the main staple food of some millions of people in Mysore State and the Deccan plateau -- is of special importance in view of its extremely high calcium content of 0.3 percent as compared with other cereals ranging from 0.01 percent to 0.07 percent (1). Recent metabolic studies on Indian children have shown that, by replacing part or all of the rice in a poor vegetarian rice diet by ragi, a considerable increase in calcium retention can take place (17). Quinoa is widely cultivated on the South American plateaus and has constituted since time immemorial the basis of the diets of the Indian populations there. Its calcium content is reported to be nearly 0.12 percent (12).

Significant calcium contributions may sometimes be made by the most unexpected of food sources. For instance, studies in Ceylon and South India have shown that the apparently low calcium content of many average diets is partly offset by large amounts of calcium present as an impurity in the crude sea salt consumed by the people. The study in South India showed that the crude salt available on the market contained 0.6 percent of calcium which is apparently available to the human system (28). Considering that substantial quantities of salt are used for cooking in those areas, the intake of calcium through salt can be significant. For example, the study in Ceylon showed that as much as 200 grams of salt containing about 300 milligrams of calcium may be used by a family in a day (7).

An example of an addition of mineral calcium to the diet comes from the United Kingdom, where calcium carbonate (as *creta preparata*) is compulsorily added to flour at the rate of 14 ounces per 280 pounds of flour (or about 0.3 percent of calcium carbonate). This contributes over 20 percent of the calcium in the average British diet.

Several other countries have adopted policies of enrichment for nutritional purposes, e.g., in the Union of South Africa bread is enriched with groundnut meal, fish flour, etc. The effects of such enrichment, which will vary with the products used, should always be taken into account.

Another example of calcium addition is the practice of many indigenous populations in Mexico and Central America of making maize tortillas. Maize diets, like most diets consisting mainly of cereals, are usually poor in calcium (20), but the situation is quite different when maize is made into tortillas. During the initial stage

of preparation, maize grain is treated with lime water before being ground. Thus, a considerable amount of calcium is introduced in the tortillas which may sometimes contain as much as 0.5 percent of calcium (23). It has been found that the average Mexican diet, for instance, may contain over 500 milligrams of calcium contributed by tortillas (9). In such cases, the total calcium intake may virtually be determined by that of the maize tortillas owing to the very low consumption of other calcium-rich foods.

Apart from the calcium which may be consumed through main foods occurring in habitual diets, considerable amounts of calcium might be ingested through peculiar local habits such as chewing of betel-leaves in India. It has been shown experimentally that calcium ingested in the process of betel-leaf chewing with lime, etc., not only increased the intake of calcium but was also retained by the subjects to a considerable extent. In fact, the amount and the utilization of calcium ingested with six betel-leaves was similar to that from 10 ounces (280 grams) of cow's milk (3). The value of chewing betel-leaves, however, has subsequently been attributed to lime added to the leaves and not to its natural mineral content (16).

Calcium will also be added to the intakes of some individuals by the consumption of pharmaceutical products. It is also probable that other unusual food habits or food practices exist in other parts of the world and make similar unexpected contributions to the dietary intake of calcium. Sufficient examples, however, have been given to make it clear that, if the calcium content of the diet is to be used as a basis for investigations of the Sr-90 content of the diet, it is important to obtain accurate information on food consumption, probably by means of household food consumption surveys, and on the calcium content of foods consumed, for which it may be necessary to analyse foods in the form in which they are consumed and not as they are produced and sold.

In general, data provided by food consumption surveys are to be preferred to those provided by food balance sheets, because surveys take into account the differences in food consumption patterns in different groups of a population, the effects of commercial food processing and, if designed to do so, the effects of food preparation in the home. These factors cannot be taken into account in the food balance sheets. Nevertheless, since large areas are involved in Sr-90 fallout, food balance sheets are likely to provide more useful

information than small-scale surveys, which are not representative of any considerable sections of the population. It is often necessary, however, to use the results of even such limited surveys since no other source of food consumption data may be available but, in such cases, the limitations of the available data should be fully recognized.

Effects of food processing

The nutritive composition of food is affected when it is treated by any kind of process, either of an industrial or a domestic nature. In the context of determining the contents of calcium, stable strontium and Sr-90 in foods, the most important processes to consider are those in which parts of the food material are removed, as in milling of cereals, or in the removal of the outside parts of vegetables before cooking, and those in which the material is treated with water, in which some of the salts of calcium and strontium are soluble, and the water is then discarded.

Several experiments illustrate the effects of milling wheat and rice. Milling the kernels of Thatcher wheat, grown in soil to which Sr-90 had been added, produced flours which contained 9 to 16 percent; the shorts, 29 to 31 percent and the bran, 53 to 62 percent of the total activity (19). The Sr-90 concentrations were almost directly proportional to the total ash in the various milling products. Similar results are reported from the United Kingdom Monitoring Survey (2). Analyses of flour prepared from grain grown in the United Kingdom showed striking differences in composition between grain, fine offal and bran. Bran gave on an average seven times the ratio of Sr-90 to calcium in flour, with an intermediate value for fine offal. Distribution was similar in imported grains. From Japan it has been reported that milling lowland rice, grown on soil which had been treated with Sr-90, produced polished rice, which contained 32 percent of the total calcium and 30 percent of the total Sr-90 of the hulled rice, and bran which contained the remainder, i.e., 68 percent of the total calcium and 70 percent of the total Sr-90 (30). For both wheat and rice differences in milling techniques alter the proportions of both Sr-90 and calcium found in the various fractions of the grain.

Some of the samples of potatoes and cabbages examined in the

United Kingdom Monitoring Survey were analyzed to show the differences in composition between the peel and the flesh of potatoes and the inner and outer leaves of cabbage. The Sr-90 content per unit weight of potato peelings was considerably higher than that of the flesh but the ratio of Sr-90 to calcium was lower. However, since the peelings account for only about 5 percent of the weight of the potatoes, the differences between the peelings and the inner flesh were not so great as to suggest that the method of preparing potatoes would have any important bearing on the Sr-90 content of the diet. The Sr-90 content per unit weight of the outer leaves of cabbages was about six times that of the inner leaves, but the ratios of Sr-90 to calcium differed much less, as the calcium content was greater in the old leaves. These older outer leaves also showed considerably higher ratios of Sr-89 to Sr-90 than the inside leaves, an indication that recent fallout made a greater contribution to the exposed leaves than to the rest of the plant. It would be very valuable to obtain similar information from other countries.

No data appear to be available on the effect on Sr-90 contents of cooking vegetables in water or otherwise but these deserve to be studied. Reference has already been made to certain cooking processes in which the calcium content is actually raised, e.g., preparation of maize tortillas.

Use of food consumption data to devise monitoring systems for Sr-90

It has already been stated that food consumption data may be used as a framework for developing systems of collection of foods for analysis for Sr-90, the ultimate object of such a survey being, of course, to estimate the dose which the human body will receive. To do this it is desirable to have available:

1. accurate estimates of total food and fluid consumption by a whole population or by sections of it, classified probably on a geographical basis;
2. actual analyses for calcium, stable strontium and Sr-90 of representative samples of foods judged to be the most important sources of these elements, and also of samples of drinking water.

From these two sets of data can be calculated the mean Sr-90 content of the total diet and also the mean ratios of Sr-90 to calcium and, if desired and possible, also to stable strontium, on which, for the purposes of biological research, though not for the assessment of hazard, it is useful to obtain data. It is important to appreciate that it is the composition of the total diet, as eaten, and not merely that of individual foodstuffs, that determines the effective dose of radiation to the skeleton. Thus, it is not really useful to make isolated analyses of foodstuffs without paying consideration to their role in the diet as a whole. It is suggested that absolute levels of Sr-90 and calcium should be determined, so that ratios can be calculated as required. Examples are given below from two countries, the United Kingdom and Japan, of means which have been adopted for obtaining this information.

UNITED KINGDOM

An example of such use of food consumption data is the 1958 survey of Sr-90 in human diet in the United Kingdom. In this survey, the main animal and vegetable sources of calcium, excluding the calcium carbonate added to flour, were identified from the records of the National Food Survey and were used as preliminary indicators of the foods to be sampled for analysis. Full information on the methods adopted are given in *Strontium 90 in Human Diet in the United Kingdom, 1958*, Agricultural Research Council Radiobiological Laboratory Report No. 1 (2).

In brief, the method was as follows. As about three-quarters of the calcium supply from natural sources came from milk and its products and as its analyses (4) had previously indicated these foods to be the most important sources of Sr-90 in the British diet, the major analytical effort was in the first instance devoted to milk. Analyses for home-produced cheese were derived from the appropriate milk analyses and, as no imported cheeses were analyzed in 1958, were also applied to imported cheese.

Vegetables were next in importance as sources of calcium. Sampling schemes were therefore devised for potatoes, various kinds of cabbages and cauliflowers, and Brussels sprouts. Although potatoes and leafy brassica are types of vegetables very commonly eaten in the United Kingdom, they are not representative of all vegetables and fruits. However, since plants do not seem to differ greatly

in the manner which they discriminate between strontium and calcium (8) it was assumed in the calculations of the Sr-90 content of the diet that the ratio of Sr-90 to calcium in potatoes is representative of that in all vegetables with subterranean storage organs, and that the ratio in leafy brassicas is representative of that in other vegetables and fruits.

As calcium of mineral origin (*creta praeparata*) is added to flour in the United Kingdom and it would not have been easy to obtain commercial samples for analysis which had not been so fortified, grain was specially milled for the survey. Flour of 70 percent extraction, bran and fine offal were prepared separately from grain grown in the United Kingdom and from that imported from abroad.

No analyses were made on samples of meat, eggs or fish. It has been shown (8) that the ratios of Sr-90 to calcium in the blood and milk of animals are, respectively, approximately 0.25 and 0.12 times the ratio in their diet. Thus, if animals slaughtered to provide meat have been grazed on pastures similar to those grazed by dairy cattle, the ratio of Sr-90 to calcium in their blood, and hence in their flesh, can be obtained by multiplying the value in milk by two. Such a calculation was used in determining the total Sr-90 content between home-produced and imported meat. Since the southern hemisphere is the major source of meat imported into the United Kingdom, and the deposition of Sr-90 is lower in that hemisphere, the general level in meat is likely to have been somewhat overestimated. Estimates based on unpublished information were made for fish and eggs, which are not important in the context of the consumption of either calcium or strontium-90 in the United Kingdom.

JAPAN

In Japan, two other different methods have been used. According to one, foods in the form in which they are eaten were collected from families and the total ash was determined in the amounts of these foods which would be eaten by a family in a month according to the National Institute of Nutrition's "standard recipes" for complete nutrition. The strontium fraction was separated from the calcium and the Sr-90 in it assayed. To compare with this estimate, a second exercise was performed. In this the results of the Na-

tional Nutrition Survey of Japan, which is conducted four times a year, were used to determine the quantities of foods consumed by different sections of the population. These were analyzed for radioactive materials and trace elements, and the results were added up to give totals for calcium and Sr-90 and other radioactive nuclides.

Sampling of foods

In any monitoring system a method of sampling foods for analysis must be devised. To obtain an estimate of the composition, in terms of any isotope, of foods consumed by the population it would seem that the point of collection of the samples should be

TABLE 9. — CALCULATION OF MEAN CONTENT OF SR-90 IN HUMAN DIET IN THE UNITED KINGDOM, 1958

Food	Calcium content in mean diet of population mg/day	Mean ratio Sr-90/Ca $\mu\mu\text{c/g}$	Estimated Sr-90 ingested $\mu\mu\text{c/day}$
Milk and cream	496	7.0	3.47
Cheese	95	7.7	0.73
Root vegetables	25	14.2	0.35
Leaf and other vegetables	38	8.7	0.33
Fruit	24	8.7	0.21
Flour and cereals of all types	47	15.9	0.75
Eggs	19	2	0.04
Meat	19	14	0.27
Fish	14	< 1	< 0.01
Other sources	4	—	—
Total animal and vegetable sources	781		6.16
Mineral sources			
(a) <i>Creta preparata</i> added to flour	243		—
(b) Water	60		0.23
TOTAL	1,084		6.39

Hence the Sr-90/Ca ratio in the total diet is about $6 \mu\mu\text{c Sr-90/g. Ca}$

the shop or the home. This method was considered in the United Kingdom, but was not adopted because of the complexity of the system of food distribution throughout the country. For example, milk is frequently moved over long distances to the main centers of consumption, the pattern of distribution is complex, and it changes during the year. Similarly, vegetables follow a tortuous, and often irregular, route from field to home. Few families live on local produce. It appeared that no simple scheme for sampling milk or vegetables at the point of consumption could be devised that would cover adequately the country's total production. The alternative of sampling somewhere near the point of production was therefore adopted.

Other systems have been adopted in other countries. For example, in the New York survey, bottles of milk were bought, as if by a housewife, for analysis. In Japan, where the methods used for food processing and preparation are believed to have an important effect on the amount of radioactive materials in the diet, the food for analysis were collected in quantities recommended in the "standard recipes" of the National Institute of Nutrition from the homes of people associated with the laboratory in which the analyses were made.

It is suggested that for adequate sampling it is of great importance to seek statistical advice and guidance on estimates of food consumption before any system of monitoring is planned.

Results of surveys on the Sr-90 content of food supplies

Few surveys have been conducted on a national basis. The most notable exception is that made in 1958 in the United Kingdom, the sampling system for which has already been described briefly. The results of this survey are summarized in Table 9.

Several estimates of the content of Sr-90 in United States diets have been made, one of which is illustrated in Table 9. The calculations shown are derived from those made by J.H. Harley (14).

For comparison with the ratio of calcium to Sr-90 shown in Table 10, two other estimates will be quoted. An unpublished estimate for 1958 for Cincinnati, which is based on local analyses for calcium (27) gives a ratio of about 12 μg Sr-90/g. Ca. A calculation based on comparison between milk and total diet gives a very rough estimate of about 11 for the whole country in 1958 (25).

TABLE 10. - ESTIMATED DIETARY SR-90 CONTRIBUTIONS - 1958 MEAN IN HUMAN DIET IN THE UNITED STATES OF AMERICA

Food	Annual quantity ¹	μmc Sr-90 per unit	Total μmc Sr-90
Milk products	<i>quarts</i> 233	8	1,860
Cereal and bakery products	<i>kilograms</i> 90	² 10	900
Meat, etc.	98	1	100
Potatoes	45	6	270
Fruits and vegetables	193	6	1,160
			4,290

Average calcium intake calculated on an annual basis = 370 g. per head. Hence the Sr-90/Ca ratio in the total diet is estimated to be about 12 μmc Sr-90/g. Ca.

NOTES: ¹ Quantities for an average household from *United States Department of Agriculture Household Food Consumption Survey, 1955*. — ² Estimated according to the milling fractions of wheat given in *United States Atomic Energy Commission Report HASL 69*, p. 128, October 1, 1959. This value and the others in this column, can be considered as only approximately representative for the United States.

Estimates have also been made for Japan, which indicate that the ratio of Sr-90 to calcium in September 1959 ranged from 11 to 15 in different parts of the country compared with a range for similar parts of the country of 1 to 4 in 1957.

Comparison of values for milk

Sufficient analyses of milk have been made in various countries to warrant a rough comparison of Sr-90/Ca ratios in 1958. This is given in Table 11.

In addition, the Lamont Geological Observatory has collected through the auspices of the Nestlé Company samples of milk representing quarterly production from many countries. Averages calculated in the observatory suggest that the ratio for Western Europe in 1957 was about 7, while those for Australia were just over 3.

TABLE 11. — MEAN SR-90/CA IN MILK IN 1958

Location	Mean μc Sr-90/g. Ca
Canada ¹	11.6
United Kingdom ²	7.0
United States of America ³	8.0

NOTES: ¹ Canadian Department of National Health and Welfare CNHW (RP-2), April 1959. — ² ARC Radiobiological Laboratory Report No. 1. — ³ Lamont Geological Observatory 1959 Annual Report.

Classification of diets

In many discussions hitherto the diets of the world have been classified into "milk" and "nonmilk" types. As the results of surveys made in many parts of the world have accumulated, it has become apparent that the ratio of Sr-90 to calcium in milk is often not lower than in many vegetable foods. The content of milk in the diet may, therefore, not be the best basis for a preliminary classification. In view of the relatively high ratios of Sr-90 to calcium in cereals, especially if they are not highly refined, it may be better to consider diets in terms of the fraction of total calcium which comes from cereal sources. Some support for such a suggestion is given by the ratios for some countries shown in Table 12. It should be noted that these countries do not include any who obtain large proportions of their calcium from starchy roots.

TABLE 12. — COMPARISONS OF RATIO OF SR-90 TO CALCIUM IN DIFFERENT FOODS — RESULTS OF SURVEYS

Country	Year	Ref.	Milk	Grain		Vegetables leaf root
				unmilled or partially milled (a)	milled (b)	
..... μc Sr-90/gram Ca						
Germany (Kiel)	1957	26	6.7	75-128	31	7-20 12-40
Japan	1956	15	2.1-2.7	81-250	36-62	1.1-2.3
United Kingdom	1958	2	7	51(c)	15.9	8.7 14
United States	1958	14	8	100(d)	50(d)	n.a. 12

NOTES: (a) including brown rice; (b) including white rice and flour; (c) 1957 crop; (d) derived from data given in Table 5 and references cited thereunder.

Cesium-137

Cs-137 and Sr-90 have similar precursors, half-lives and fission yields, but different chemical properties, which make their behaviors in food chains and in the body different. The following is an extract from the 1958 United Nations Committee Report:

“Cs-137 concentrations are often expressed by the Cs-137/potassium ratio. Some evidence exists, however, that the metabolism and routes of entry into the human body of these two elements are to some degree different. For example, in man the biological half-life of potassium (35 days) is apparently shorter than that of cesium. An analogy of Sr-90/calcium ratios should therefore not be implied.”

This suggests that the potassium content of the diet may not necessarily be the most useful guide to the analysis of foods for Cs-137, and even if it were thought to be a useful guide, there would be difficulty in using it. The potassium content of food supplies is very rarely calculated for normal diets and most tables of food composition do not give analyses for potassium.

However, it would probably be more useful to base future monitoring work on the analyses of foods for Cs-137 that have already been made.

Results of surveys on the Cs-137 content of food supplies

In the spring of 1956, the Los Alamos Scientific Laboratory began an extensive survey of the ratio of Cs-137 to potassium in the United States' milk supply. Analyses were made on samples of dried or fresh milk from 11 stations in various sections of the United States, a number which was too small to give adequate coverage for the national milk supply. Therefore, during 1957, dried milk from 33 places was sampled on a routine basis and in addition a few spot checks were made on dried milk samples from other countries. For about half the year monthly samples were collected independently from five milksheds for analysis by the United States Public Health Service. Both series of analyses continued in 1958. The results obtained are given in Table 13 (18).

TABLE 13. - AVERAGE Cs-137 CONTENT OF UNITED STATES MILK

Year	Los Alamos Scientific Laboratory	United States Public Health Service
 $\mu\mu\text{c}$ Cs-137 per	gram potassium
1956	24	-
1957	32	34
1958	42	44

Five samples from Argentina gave an average of 9 $\mu\mu\text{c}$ of Cs-137 per gram of potassium, and milk analyzed by workers in several other countries during 1956-57, including Japan, Norway, and Sweden, was found to have a concentration ranging from 20 to 90 $\mu\mu\text{c}$ of Cs-137 per gram of potassium (22). Calculations have been made which suggest that in the United States about 60 percent of the body Cs-137 comes from milk, 25 percent from meat, 7 percent from flour and cereals, 5 percent from vegetables and 3 percent from citrus fruits (18). A recent study made at the Atomic Energy Research Establishment at Harwell, United Kingdom, suggests that in certain individuals surveyed milk was responsible for slightly under one half of the average Cs-137 burden, and that meat, fruit and potatoes are other sources to be considered (24). Results from Germany (20) indicate that ratios of Cs-137 to potassium in potatoes may be up to one fifth of those in milk; thus they also suggest that when potatoes are an important item in the diet they may be one of the main dietary sources of Cs-137. No estimates appear to have been made of the major sources of body Cs-137 in diets where milk does not constitute a major item, but measurements of rice in Japan in 1956-57 showed a concentration of about 5 $\mu\mu\text{c}$ Cs-137 per gram of potassium (22). It will be noted that all the results presented here are given in terms of the ratio of Cs-137 to potassium.

As there is no close relationship between the biological behavior of the two isotopes, it is suggested here that whereas it is desirable to analyze materials, particularly milk and human tissues, for both it will be more useful to express results in terms of the composition of foods, i.e., to express both per kilogram of the wet weight of foods.

Iodine-131

I-131 apparently will be important only after accidental release or at short times after production and dispersal. The experience of the Windscale accident suggests that fresh milk is the only important dietary source of I-131. The half-life of the isotope is sufficiently short to make any products manufactured from contaminated milk unimportant as sources of I-131. As green vegetables are usually eaten as fresh as possible, it may be worthwhile to investigate the extent to which I-131 is removed during the preparation and cooking of such vegetables. There is also a possibility that under such emergency conditions the contamination of eggs from hens on free range might also be significant.

The Windscale accident, of course, provides evidence only for a population eating a British type of diet. Nevertheless, it may be worthwhile to record that experience from that accident leads to the conclusions that the quantity of I-131 deposited in the thyroid will depend primarily on the amount of dairy produce consumed and the length of time between production and consumption. Another important factor is the content of stable iodine in the diet, which has an influence on the proportion of ingested iodine which is transferred to the thyroid.

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GENERAL SOURCES OF NATURAL CALCIUM IN THE FOOD SUPPLIES OF SELECTED COUNTRIES

[Expressed as percentages of estimated total calcium content. Calculations based on food balance sheets, 1954-1956 average, FAO, 1958)

Country	Cereals	Starchy roots	Pulses, nuts	Veg- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	Total supply of Ca (expressed in mg/head/day)
Europe										
<i>Austria</i>										
% of total Ca supply	6	2	1	10	3	1	1	—	76	900 100
<i>Denmark</i>										
% of total Ca supply	7	3	1	9	2	1	1	1	76	950 101
<i>Finland</i>										
% of total Ca supply	6	2	—	2	1	1	1	1	87	1 350 101
<i>Germany</i>										
% of total Ca supply	6	4	1	7	3	1	1	1	76	850 100
<i>Italy</i>										
% of total Ca supply	10	1	4	22	4	1	2	1	57	600 102
<i>Norway</i>										
% of total Ca supply	6	2	1	4	1	1	1	2	82	1 150 100

¹ I.e., excluding additions of calcium salts of mineral origin to flour or other foods.

ANNEX A

CONTRIBUTIONS OF NATURAL CALCIUM IN THE FOOD SUPPLIES OF SELECTED COUNTRIES (continued)
Expressed as percentages of estimated total calcium content. Calculations based on food balance sheets,
1954-1956 average, FAO, 1958)

Country	Cereals	Starchy foods	Pulses, feed	Vege- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	Total supply of Ca (expressed in mg/head/day)
<i>Sweden</i>										
% of total Ca supply	4	2	1	3	2	1	1	1	85	1 050 100
<i>United Kingdom</i>										
% of total Ca supply	5	2	2	10	2	2	2	1	76	850 102
<i>NORTH AMERICA</i>										
<i>Canada</i>										
% of total Ca supply	3	1	1	4	2	2	2	1	83	1 050 99
<i>United States</i>										
% of total Ca supply	3	1	2	12	2	2	2	—	77	1 150 101
<i>LATIN AMERICA</i>										
<i>Argentina</i>										
% of total Ca supply	6	4	3	14	3	4	1	—	64	700 99

e, excluding additions of calcium salts of mineral origin to flour or other foods.

DIETARY SOURCES OF NATURAL CALCIUM IN THE FOOD SUPPLIES OF SELECTED COUNTRIES (continued)
 (Expressed as percentages of estimated total calcium content. Calculations based on food balance sheets,
 1954-1956 average, FAO, 1958)

Country	Cereals	Starchy Roots	Pulses, nuts	Vege- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	Total supply of Ca (expressed in mg/head/day)
<i>Chile</i>										
% of total Ca supply	26	4	6	27	3	2	1	22	8	350 99
<i>Guatemala</i>										
% of total Ca supply	6	1	1	7	2	3	1	—	78	700 99
NEAR EAST AND AFRICA										
<i>Egypt</i>										
% of total Ca supply	20	1	6	20	11	1	—	1	40	400 100
<i>Israel</i>										
% of total Ca supply	10	1	4	18	6	1	2	1	57	900 100
<i>Turkey</i>										
% of total Ca supply	27	1	7	24	7	1	—	—	33	450 100
<i>Union of South Africa</i>										
% of total Ca supply	13	1	2	13	4	2	1	1	63	400 100

* i.e., excluding additions of calcium salts of mineral origin to flour or other foods.

ANNEX A

GENERAL SOURCES OF NATURAL¹ CALCIUM IN THE FOOD SUPPLIES OF SELECTED COUNTRIES (*concluded*)
(Expressed as percentages of estimated total calcium content. Calculations based on food balance sheets,
1954-1956 average, FAO, 1958)

Country	Cereals	Starchy roots	Pulses, nuts	Vegetables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	Total supply of Ca (expressed in mg/head/day)
ASIA AND FAR EAST										
<i>Ceylon</i>										200
% of total Ca supply	20	9	13	30	—	—	1	4	23	100
<i>India</i>										300
% of total Ca supply	21	2	24	7	2	—	—	—	44	100
<i>Japan</i>										350
% of total Ca supply	14	7	24	26	1	—	1	15	11	99
<i>Pakistan</i>										350
% of total Ca supply	18	—	7	7	4	—	—	—	63	99
OCEANIA										
<i>Australia</i>										850
% of total Ca supply	5	1	1	10	5	3	1	—	73	99
<i>New Zealand</i>										1 100
% of total Ca supply	4	1	1	9	2	2	1	—	79	99

¹ I.e., excluding additions of calcium salts of mineral origin to flour or other foods.

SOURCES OF CALCIUM IN THE PER CAPUT FOOD SUPPLIES OF SELECTED COUNTRIES
(Calculations based on dietary surveys)

Country	Cereals	Star- chy roots	Pulses, nuts	Vege- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	TOTAL Ca	No. persons	Source
EUROPE												
<i>United Kingdom</i> ¹ mg. Ca/cap/day % Ca of total supply	290 28%	25 2%		46 4%	16 2%	23 2%	19 2%	14 1%	591 58%	1 024 99%	country wide	Agric. Research Council Radiobi- ological Laboratory Report No 1, <i>Stron- tium 90 in Human Diet in the United Kingdom 1958</i> . London, H.M.S.O. 1959.
NORTH AMERICA												
<i>United States</i> <i>Urban</i> mg. Ca/cap/day % Ca of total supply	167 15%	11 1%	16 2%	81 7%	37 3%	37 3%	24 2%	incl. meat	716 64%	1 111 ² 99%	6 000 house- hold	<i>Dietary Levels of Households in the U.S.</i> Household Food Consumption Surveys 1955, Re- port No 6. U.S. De- partment of Agri- culture, Washing- ton, D.C. for all U.S. data, here and on page 125
<i>Rural nonfarm</i> mg. Ca/cap/day % Ca of total supply	212 18%	12 1%	24 2%	74 6%	32 3%	34 3%	26 2%	" "	722 62%	1 160 ² 99%	"	

¹ 243 mg. of *creta praeparata* added to the cereals.² Misc. foodstuffs included in total Ca figure.

SOURCES OF CALCIUM IN THE PRE-CAPIT FOOD SUPPLIES OF SELECTED COUNTRIES (continued)
(Calculations based on dietary surveys)

Country	Cereals	Starchy roots	Pulses, nuts	Veg- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	TOTAL Ca	No. persons	Source
<i>For Japan</i>												
mg. Ca/cap/day	222	14	25	79	30	33	32	incl.	855	1 320 ¹	house-	
Ca of total supply	17%	1%	2%	6%	2%	3%	2%	meat	65%	100%	hold	
<i>Latin America</i>												
Venezuela												
mg. Ca/cap/day	59	12	71	37	12	17	and milk prods. 172		433	865	769	González S., Una
Ca of total supply	7%	1%	8%	4%	1%	2%	20%		50%	99%	persons	encuesta alimenta- ria en las familias. Arch. Venez. de Na- trición, Vol. VII, No. 2, 1956 p. 167
<i>NEAR EAST AND AFRICA</i>												
<i>Ivory Coast</i>												
Village												
mg. Ca/cap/day	3	271	14	12	3	14		419	—	736		Territoire de la
Ca of total supply	—	37%	2%	2%		2%		57%		100%		Côte d'Ivoire - En- quête Nutrition -
Camp												Niveau de Vie, Sub- division de Bou- gouan, 1955-56
mg. Ca/cap/day	1	336	2	28	7	27		389		790	7 254	persons
Ca of total supply	—	43%	—	4%	1%	3%		49%		100%		

¹ Misc. foodstuffs included in total Ca figure.

SOURCES OF CALCIUM IN THE PER CAPIT FOOD SUPPLIES OF SELECTED COUNTRIES (continued)
(Calculations based on dietary surveys)

Country	Cereals	Star- chyl roots	Pulses, nuts	Vege- tables	Fruits	Meat, poultry	Eggs	Fish	Milk, cheese	TOTAL Ca	No. persons	Source
ASIA AND FAR EAST												
India												
1947 Ca/cap/day	128		76	80	2		5		160	4691	83 692	Indian Council of Medical Research, Special Report Series No. 25. A Supplement to the Results of Diet Surveys in India 1935-1948, by K. Mitra, New Delhi 1953
% Ca of total supply	27%		16%	17%	—		1%		34%	99%	persons	
Japan												
1947 Ca/cap/day	53	9	101	64	9	1	8	55	34	3841	Nation wide	Ministry of Health and Welfare, Japan. Nutrition in Japan, 1953.
% Ca of total supply	14%	2%	26%	17%	2%	—	2%	14%	9%	99%		

Misc. foodstuffs included in total Ca figure.

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